ISOTOPES AND RADIATION TECHNOLOGY

A Quarterly Technical Progress Review

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FEATURE ARTICLE:

Radioisotopes and Radiation Developments in the USA—1967

By E. E. Fowler

TECHNICAL PROGRESS REVIEWS

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Isotopes and Radiation Technology

is a Technical Progress Review prepared by the Isotopes Information Center, Oak Ridge National Laboratory, at the request of the Divisions of Technical Information and Isotopes Development, U. S. Atomic Energy Commission. This Review is intended to assist those interested in keeping abreast of significant developments in the fields of isotopes and radiation technology. It is not a comprehensive review of all literature published in this field during a given quarter; rather, it is a mechanism for presenting concise, selected reviews of information on subjects of prevailing Commission interest as it becomes available.

This Review attempts to relate the published results of research and development sponsored by the Division of Isotopes Development to significant developments in radioisotopes and radiation technology, as reported in the world literature. Coverage includes isotope production and development, isotope technology development, isotope applications technology, process radiation development, and radiation processing of food and medical supplies.

In addition to reviews of current literature and special review articles dealing with specific isotopes, facilities, and applications, this publication occasionally contains feature articles prepared by recognized experts on specific topics of current interest. Critical evaluations and interpretations presented are those of the editors and invited reviewers; therefore readers are encouraged to consult the original references in order to obtain all the background of the work reported and the interpretation of the results given by the original authors.

The subject headings listed below have been adopted to help maintain continuity and order, from one issue to the next, in the material reviewed. All reviews, except Feature Articles, will be arranged under these headings; but any one issue will not necessarily contain all the headings.

Isotope Production and Development
Isotope Technology Development
Food Irradiation
Process Radiation Development
Source Development
Isotopic Heat and Power Development
Applications in Life Sciences
Safety
Isotopes Around the World
Miscellaneous

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FEATURE ARTICLE

Radioisotopes and Radiation Developments in the USA—1967*

By E. E. Fowler†

Abstract: A general survey is given of the U. S. program on radioisotopes and radiation developments in 1967. Three main phases of the program are presented: (1) development, as well as present emphasis and future trends, of the U. S. Atomic Energy Commission's research and development program in isotopes and radiation technology; (2) status and trends in current uses of isotopes, both by industry and government; and (3) current status of production and sales of radioisotopes and radiation sources.

The U. S. program on radioisotopes and radiation development is designed and conducted as a broadly based multidisciplined technological effort. Its objective is to develop technology and demonstrate applications of radioisotopes and radiation important to the national economy and to stimulate independent research and development by other government departments, industry, and the scientific community. Through this focusing of our total national technical and industrial capabilities, we expect to realize full achievement of the potential contributions of radioisotopes and radiation to our national goals, economy, and welfare.

I will discuss in this paper three main phases of the radioisotopes and radiation program in the United States: (1) development, as well as present emphasis and future trends, of the U. S. Atomic Energy Commission's research and development program in isotopes and radiation technology; (2) status and trends in current uses of isotopes, both by industry and government; and (3) current status of production and sales of radioisotopes and radiation sources. My discussion will provide a general survey of progress and activities in the United States since other speakers from my country will cover specific technical details on particular programs.

*Presented at the 8th Japan Conference on Radioisotopes, Tokyo, Japan, Nov. 13–16, 1967.

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USAEC Isotopes and Radiation Development Program

The current USAEC Isotopes and Radiation Development Program is broadly based, with major research and development work being directed to environmental and pollution studies, analysis and process control, food preservation, process radiation, and thermal- and electric-power applications.

In addition to these programs, which will be reviewed below, the USAEC has also sponsored a program to study the safety of sealed sources produced both commercially and in Commission laboratories. This work has resulted in the development of a method for classification of sealed sources, which is based on the ability of the source to withstand heat and mechanical shock.[1] This classification system is under study for use as a uniform method for describing the integrity of source capsules. The USAEC has also established an Isotopes Information Center at its Oak Ridge National Laboratory to collect, evaluate, and disseminate information on radioisotopes.[2] The Center uses scientific and technical publications reported throughout the world.

Environmental and Pollution Studies

In the United States, over 142 million tons of noxious gases and particulate matter is introduced into the air each year. This very nearly equals the 160 million tons of solid waste that Americans generate each year. In New York City, each square mile is covered by 60 tons of soot each month.[3] While the problems of air and water pollution have been with us for many years, greatly increased concern and attention is now being directed to this national problem.

When President Johnson signed an Executive Order on control of air pollution in federal facilities, he em-

phasized that, although there were great technological and economic problems in the abatement of air pollution, the battle for cleaner air remained a major objective of his administration and an essential element in a better environment for America. The U. S. Department of Health, Education, and Welfare has the primary responsibility for solving air-pollution problems; the USAEC is cooperating by developing nuclear methods to assist in studying these problems.

Over the years the USAEC has sponsored a number of research programs to develop nuclear techniques for pollution studies. These studies have resulted in the development of (1) several instruments and analytical methods for determining SO_2 in air,[4–6] (2) an instrument for determining ozone in air,[7,8] (3) a gage for measuring the amount of suspended solids in a flowing stream,[9] and (4) a method using ^{51}Cr to determine the point when ion exchangers used to remove chromium and other metals from a plating bath are exhausted[10] and thus are no longer effective in clearing up solutions that are to be dumped into streams.

Sulfur Dioxide Pollution

One of the most serious air-pollution problems in the United States today involves sulfur dioxide. Significant amounts of SO_2 are introduced into our atmosphere primarily through the burning of fossil fuels (the sulfur content of bituminous coal in the United States ranges from 0.5 to 6%). A 150-Mw(e) utility plant burns 75 tons of coal per hour. With each ton of coal burned, 400 thousand cu ft of total gas, containing about 0.3 wt.% SO_2, is produced. If regulatory limits are established for discharge of sulfur dioxide into the atmosphere, effective control could be maintained either by selection of fuels with low sulfur content or by scrubbing of the off-gases to remove the sulfur dioxide. The use of either of these techniques will require improved analytical methods.

Brookhaven National Laboratory has initiated work on a method to trace sulfur dioxide in stack gases by using the variations in $^{32}S/^{34}S$ ratio that occur in natural fossil fuels.[11] Feasibility studies completed by them have shown that the variation in this ratio from fuel to fuel may be large enough for it to be used in identifying the sulfur dioxide coming from a particular stack. Thus the sulfur dioxide would act as its own tracer, and the variation in $^{32}S/^{34}S$ ratio should be a very valuable aid in studying air pollution from this source if an adequate technique can be developed. It is possible that an enriched stable sulfur isotope or an easily activable element such as that used by Nakasa and Ohno[12] of the Central Research Institute of Electric Power Industry, Tokyo, will have to be added to the fuel, but the variation in the natural ratio promises to be sufficient so that tracing without adding other tracer materials may be possible.

With the isotopic ratio tracer method, many valuable studies on the mechanism of air pollution should be possible. One can determine, for example, (1) the conditions under which plumes from tall stacks will or will not break through the inversion layer; (2) if the plume does not break through the inversion layer, whether the plume remains aloft or mixes with the lower atmosphere; (3) the behavior of SO_2 in the plume and the conversion of SO_2 to sulfates in the plume; and (4) the behavior of particulate sulfates from the plume. Work is continuing on these problems.

We have recently started three new projects to establish feasibility of development of better methods for SO_2 determination in stack gases. In one of these, radiation absorption and radiation scattering will be examined as the method of determination; in another, X-ray fluorescence will be examined; while all methods, both nuclear and nonnuclear, will be evaluated in the third study.

Water Pollution

The pollution of waters by industrial plants is as serious a problem in the United States as is that of air pollution. We are therefore sponsoring isotopes research to provide scientific and technical data, as well as methodology, to gain a better understanding and to assist in the solution of this important national problem.

The paper industry is typical of industries in the United States which have a problem with stream pollution. We are currently sponsoring a project at Washington State University, Pullman, Wash.,[13] to develop tracer methods to determine the origin of these wastes and how they can best be handled. The technique currently under study is to tag the wood pulp with iridium or other easily activable materials, to remove pulp samples from appropriate locations in the process stream, and to activate the samples in a reactor; the iridium or other tracers can be used to follow the pulp through the entire process and into the waste stream. This technique will also be used to study other materials in the paper-manufacturing process, such as the filler clays, titanium dioxide, alum, and sizing agents.

Another serious source of water pollution in the United States is drainage of waters from strip and deep shaft coal mines into streams and rivers. These waters are acid and contain enough ferrous and ferric sulfates from pyrites to do considerable harm to normal aquatic life in such bodies of water and to make them undesirable for recreation. In a preliminary study at Brookhaven National Laboratory, workers showed that addition of limestone and aeration, followed by a radiation-induced oxidation, would result in the neutralization of these wastes and precipitation of the iron in an easily filterable form. Studies are continuing on this problem[14] to determine economics and feasibility on a larger scale.

Sand Movement Studies

I am sure that I do not have to tell this audience that isotopes have been used for many years in the study of sand movement on beaches. Some of the best work in the world in developing these methods has been here in Japan.[15–18]

We have recently started a new study in this field in which ^{133}Xe is used as the tracer. Quartz sand can be impregnated with this isotope by heating the sand to about 920°C in a xenon atmosphere at a pressure of about 20 mm Hg (abs.). This xenon-sand tracer has the following advantages: (1) the tracer sand is hydrodynamically identical with the other sand on the beach; (2) the health hazard is negligible because of the chemical and biological inertness of xenon; (3) consecutive experiments can be run in the same location because of the short (5.3 days) half-life of ^{133}Xe; and (4) little or no shielding of the sand is needed.

The sand is introduced onto the beach in the usual way, and its movement is followed by a scintillation counter, which is housed in a perforated steel cylinder (Fig. 1) towed behind an amphibious vehicle. Tests to date have been run at Cape Kennedy, Fla., and at Point Conception, Calif. These tests were run at depths between 30 and 60 ft, and the sand movements were traced successfully for several days in each test.

Analysis and Process Control

Since 1958 the USAEC has spent over $2 million on basic and applied work with neutron activation analysis. Private industry probably has spent a like amount on this method. Much of this work has been reported at previous Japan Radioisotope Conferences.[19] The basic technology has been firmly established, and the program is now concerned mainly with applying this technique to forensic work and to continuous on-stream analysis of a number of commercial products.[20] The latter application will be reviewed by J. R. Rhodes during this conference.

In the past year, added emphasis has been given to X-ray fluorescence analysis in our program,[21] and a number of gaging applications that are useful in the space program have been studied. Some of these applications are discussed below.

X-Ray Fluorescence Analysis

We have a number of studies under way to develop practical uses of X-ray fluorescence analysis, using isotopically excited sources, in such fields as forensic science. In these studies, we hope to take advantage of the specificity of the method for detection of individual elements, the portability of the apparatus required, and the nondestructive nature of the test. In one study, for example, lead residues around suspected bullet

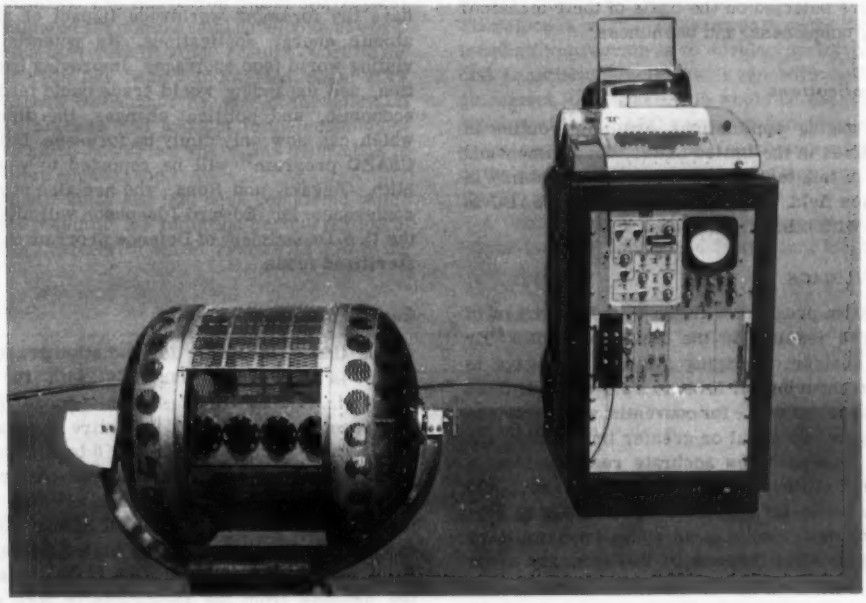

Fig. 1 Ocean-floor radiation detector. The grill is open to show the scintillation counter. The counting and recording apparatus on the right is on board ship.

holes were positively identified. A small portable instrument that can be carried into the field appears feasible. Prior to the use of X-ray fluorescence for this determination, material around the bullet hole had to be removed and transported to a laboratory for dissolution of the lead for chemical analysis.

Radioisotope-Light-Source Photometer[22]

A beta-emitting radioisotope, ^{36}Cl, has been mixed with a scintillating material to provide an unusually stable light source for use in photometric analysis. In addition to its stability, the source is very small and does not require an external electric-power source. Because of the long half-life of ^{36}Cl (3×10^5 years), the stability of the light source is determined by the radiation decomposition of the scintillator. A photometer with such a light source has an instrumental error of about $\pm 0.2\%$ and a useful life of at least 5 years before the scintillator is changed. This development was judged to be one of the 100 best technical inventions in 1966, in the annual competition sponsored by the magazine *Industrial Research*. The competition is sponsored by the magazine to recognize and to stimulate scientific and technical innovations. The 100 products were selected on the basis of their technical importance, uniqueness, and usefulness.

Gaging Applications

Although gaging applications are now routine in many industries in the United States, development and application of this technology to new areas, mainly in the aerospace field, are under study by the USAEC in cooperation with other federal departments.

AIRPLANE OIL GAGE

A cooperative program with the U. S. Department of the Air Force resulted in the development of a ^{85}Kr continuous nuclear oil gaging system. This gage is capable of measuring oil level to $\pm 2\%$, as contrasted with an accuracy of $\pm 20\%$ for conventional capacitance and float gages. Of equal or greater importance, the nuclear oil gage provides accurate readings over a large range of attitude, altitude, and acceleration and is not hindered in its operation by oil frothing. Recently an accident involving an operational military aircraft was avoided because of the accuracy of the nuclear oil gage. Twenty-five minutes after takeoff, the flight was aborted when the gage showed only 14 pints of oil remaining. Upon landing, inspection of the engine revealed a leaking oil line coupling. If the aircraft had remained in flight 15 min longer, it would have completely depleted its oil supply.

ZERO-GRAVITY PROPELLANT GAGE[23]

A propellant gage that works under zero gravity has recently been developed. This gage is particularly interesting because it is based on the principle of isotope dilution rather than on the usual radiation absorption technique. A small quantity of ^{85}Kr gas is injected into the ullage space of the propellant-filled tank. As propellant is expelled from the tank, the ullage-space pressure is maintained by a regulated influx of nonradioactive pressurizing gas. This inflowing gas dilutes the concentration of the radiotracer gas, which is continuously measured with radiation detectors. The pressurizing gas within the tank's ullage volume forces propellant expulsion. The count rate from the detectors, being proportional to the ^{85}Kr concentration, is related, by means of a simple algebraic equation, to the volume of propellant remaining. The count-rate data can be converted into a digital quantity readout or used as input for propellant management. In tests the gaging error was consistently less than 0.5% over the entire range of propellant volume.

Radiation Preservation of Foods

The use of radiation to pasteurize or sterilize food is perhaps the largest potential use of ionizing radiation. More importantly, this technology promises to have the foremost worldwide impact of all peaceful atomic energy applications. Its potential for alleviating world food shortages, improving human nutrition, and expanding world trade could lead to social, economic, and political changes, the dimensions of which can now only dimly be foreseen. Details of the USAEC program[24] will be reported to you by Goldblith, Otagaki, and Ross, who are also attending this conference. Dr. Edward Josephson will also report on the U. S. Department of Defense program on radiation-sterilized foods.

Current Program

The USAEC program for radiation preservation of foods is concerned primarily with the radiation pasteurization and disinfestation of foods. The USAEC has recently reevaluated its entire program in this field. The objective is to select 6 to 10 food products or product groups that demonstrate the greatest opportunity for commercialization. Before a process for irradiating food for general huma consumption can be commercialized in the United States, the U. S. Food and Drug Administration (FDA) must approve the process from the viewpoints of both safety and efficacy; therefore our program is structured to obtain the data in those areas required by the FDA. Of course, the ultimate goal of the program is to provide to the consumer foods of high quality and in a fresh or nearly fresh condition.

The following general guidelines are being followed in selecting the food products for full study and development: (1) the product should be a large-volume staple; (2) radiation processing of the product should

appear technically feasible from previous work; (3) the product should be one that is subject to spoilage of a nature to create an economic or health problem; (4) the cost of spoilage would be great enough to justify cost of radiation processing; (5) the product is concentrated in sufficient volume at a single location during the processing sequence to make radiation processing feasible; (6) production of and demand for the product, perhaps in conjunction with other products in the program, should be adequate throughout the year to allow the economic use of a commercial-size radiation processing facility; (7) radiation processing, in comparison with alternative processes, would provide a clearly defined economic benefit for the product; (8) companies or organizations that have both the financial strength and managerial attitude to be receptive to new processes should exist in the industry; and (9) a significant market should exist for the product, i.e., success in commercializing radiation processing should not be contingent on major marketing effort to change consumer eating and buying habits.

In the past year a cost-benefit study[25] was completed for East Coast finfish fillets (cod, haddock, sole, and flounder), West Coast Dungeness crab, Alaskan king crab, and tropical fruits (papaya, mangoes, and bananas). The East Coast finfish and tropical fruit were selected for commercial development work. Based upon the cost-benefit study results, work on the two types of crab has been discontinued. Some work will be continued on bananas because of the potential value of this process and product to the economy of developing tropical countries where this product is of major economic importance.

A cost-benefit study has just been completed which evaluates the potential for radiation pasteurization of fresh meats. Analysis of these results is now being carried out along with technical evaluations leading to a decision on the desirability of initiating a full-scale research and development program. A decision should be made soon as to the future of this program.

The USAEC, the U. S. Department of Agriculture, and the Bureau of Commercial Fisheries of the U. S. Department of Interior have recently started a joint evaluation to provide a sound basis for selecting the remaining products for inclusion in the full technology development program. The results of this study should be available this coming year. The following products are being considered: East Coast shellfish (blue crab, oysters, clams), Gulf Coast fin- and shellfish (red snapper, mullet, shrimp, oysters), West Coast finfish (halibut, rockfish, ocean perch, sole, flounder), freshwater fish (catfish), strawberries, processed eggs, fresh chicken, rice, wheat flour and packaged flour mixes, mushrooms, tomatoes, peaches, nectarines, and oranges.

Irradiator Program

The FDA has indicated that it will not consider approval of radiation processing of any food unless a beneficial effect is shown; this may require, in particular cases, large pilot-plant-scale demonstration. Therefore a very necessary part of the USAEC food-irradiation program is to develop prototype commercial irradiation facilities. Even without the FDA requirement, these facilities would be necessary for the support of the USAEC research and development program and to demonstrate the commercial feasibility of the process. Table 1 gives the status of the current USAEC irradiation facilities.

A cooperative USAEC–Department of Defense–Department of Commerce Pilot Plant Meat Irradiator

Table 1 IRRADIATORS SUPPORTING THE FOOD-IRRADIATION EFFORT

Irradiator	Description	Location	Purpose
		Research	
Research	30,000–40,000 curies of ^{60}Co, pool type	Massachusetts Institute of Technology University of California (Davis) University of Washington University of Florida	On-site research
On-Ship No. 3	36,000 curies of ^{60}Co, 18-ton portable unit	Seattle, Wash.	Irradiation at sea
Portable Cesium No. 1	170,000 curies of ^{137}Cs, 18-ton portable unit	Various industries	Demonstration to and use by industry
		Pilot Plant	
Marine Products Development Irradiator (MPDI)	250,000 curies of ^{60}Co, 1000 lb/hr at pasteurizing dose	Gloucester, Mass.	Seafood irradiation
Mobile Gamma Irradiator (MGI)	100,000 curies of ^{60}Co, 1000 lb/hr at pasteurizing dose	Davis, Calif.	Fruit irradiation
Grain Products Irradiator (GPI)	25,000 curies of ^{60}Co, 5000 lb/hr bulk or 2800 lb/hr packaged	Savannah, Ga.	Grain products disinfestation
Hawaii Development Irradiator (HDI)	225,000 curies of ^{60}Co, 4000 lb/hr at pasteurizing dose	Honolulu	Tropical fruits

Project was initiated in July with the signing of a contract with IRRADCO, Allentown, Pa. The company—jointly owned by Teledyne, Inc.; Liggett & Myers Tobacco Co.; Martin-Marietta Corp.; and Uniroyal, Inc.—was selected from 10 firms who responded to the USAEC public solicitation for proposals. Under the terms of the contract, IRRADCO will design, construct, and operate an irradiation facility in Allentown, Pa., capable of processing 3 million lb of product per year at sterilizing doses of 5 million rads. IRRADCO's expenditures are projected at $1 million.

The USAEC is to provide $185 thousand for the purchase of a 400-kilocurie ^{60}Co source, plus limited technical assistance. The U. S. Department of Defense has agreed to purchase 300,000 lb of radiation-sterilized meat annually for the first 3 years of operation and, also, to provide limited technical assistance. In addition to the planned supply of FDA-cleared meat products to the U. S. Department of Defense, the company has commercialization plans for developing radiation-processed foods for the general consumer market in the United States. A corollary interest of IRRADCO is in radiation-processed pet food, which is also subject to FDA approval in the United States.

The USAEC is also cooperating in the development of radiation processing of foods internationally, not only by making the scientific and technical information that we are developing in our own program generally available but also in selected instances through the loan of research irradiators. Irradiators on loan or projected for loan to other countries are shown in Table 2. Generally the initial loan is for a 2-year period. Countries receiving the irradiators on loan will not only be developing their own knowledge and capabilities but will also lend important support to the U. S. efforts by providing technical information pertinent to our work. Over 50 countries now have active programs in food irradiation. We understand that 20 other countries also have research program plans under study for possible implementation.

Table 2 IRRADIATORS ON LOAN TO OTHER COUNTRIES

Unit	Country	Date of loan	Products
On-Ship No. 1	Israel	March 1966	Citrus
	Chile	June 1968	Potatoes, fish
On-Ship No. 2*	Iceland	April 1968	Fish
Portable Cesium No. 2	Argentina	Spring 1968	Bovine blood, fruit, fish
Portable Cesium No. 3	India	Spring 1968	Wheat products
Research	Pakistan	Spring 1968	Wheat products

*Formerly on the trawler *Delaware* based in Gloucester, Mass., USA.

Process Radiation

The goal of the process radiation program[26] in the United States is to stimulate radiation technology application and to help develop economically useful process radiation systems. The chemical processing industry is highly sophisticated and deals with a wide variety of products and processes; thus process radiation must compete with highly efficient existing technology that can often adapt itself to meet the competition of a new process. A radiation process must therefore demonstrate advantages over the conventional processes with at least equal or improved economies. The advantages of radiation processing are well known to those of you in the field: (1) processes can be initiated at low temperature; (2) the process initiator is external and thus no impurities are introduced; (3) processes can be initiated in the solid phase; (4) the processing may be done on the material in its final geometric form or in its final container; and (5) the process reaction rate may be instantaneously regulated by mechanical adjustment of the radiation source, which is external to the system.

The USAEC research and development program has been successful in helping to catalyze the growth of industrial process radiation in the United States. An estimated $250 million worth of products was manufactured by radiation processing in 1966. Many of these radiation-produced products have been the subject of reports by U. S. investigators at earlier Japan conferences.

The overall process radiation program consists of three complementary efforts: basic technology development, radiation engineering, and specific process applications.

Basic Technology Development

The USAEC program in basic radiation technology is aimed at determining the basic mechanism of interaction of beta and gamma rays with specific chemicals, with the objective of determining implications for applied radiation chemistry. Three major types of chemical reactions are being studied: organic chain reactions with a strong emphasis on polymerization, solid-state reactions, and surface catalysis.

The study at Brookhaven National Laboratory of the application of radiation to polyethylene production[27] has been carried to the point where at least one industrial firm, and perhaps two, is constructing a full-scale pilot plant at its own expense. The basic incentive for this is that radiation-initiated polymerization of ethylene promises to be a less costly process than the conventional chemical initiation now in use. Sales of polyethylene in the United States are so large that, if only a few percent of the total production was made by radiation processing, it would amount to a sizable saving. Safer control of the highly exothermic reaction is also possible because the external radiation source can be engineered to respond instantaneously as the reaction rate changes. One may note that this feature is common to all exothermic chain reactions.

Another potential application of radiation processing being investigated in this area is the impregnation of concrete with plastics. The objective is to determine whether materials can be produced that will be resistant to hot brine and distilled water and possibly be usable construction materials for water desalination and cross-country pipelines.

Radiation Engineering

A substantial portion of the USAEC's Process Radiation Development Program is devoted to the generation of engineering data essential to the use of radiation as an industrial process tool. Several important achievements made in this program in the past year are:

(1) At Brookhaven National Laboratory encapsulated ^{60}Co strip sources were fabricated in which the source and encapsulating materials are bonded to each other. Because of new techniques, this source, while the same overall size as earlier strip sources, contains approximately 25% additional ^{60}Co.

(2) Self-absorption by ^{60}Co and ^{137}Cs was analytically investigated and experimentally verified.[28] These data will permit users or designers of irradiation facilities to readily select appropriate sources and source configurations for optimization of irradiation efficiency and minimization of radiation costs.

(3) Depth-dose experimental comparisons of ^{137}Cs and ^{60}Co for homogeneous, finite targets were completed. From these results irradiation processors can easily determine, largely through extrapolation, actual absorbed doses in different media under the same or similar irradiation conditions.

Specific Process Applications

WOOD–POLYMER MATERIALS

Our research and development objectives on thick product-stock wood–plastics have essentially been accomplished, and we are terminating work in this area. We may, however, pursue a small research effort aimed at developing an economic process for treating veneers. Analysis of the industry has indicated that, if a large commercial use of wood–polymer materializes, it will be in the field of veneers.

MICROBIOLOGICAL INHIBITION SYSTEMS EMPLOYING RADIATION[29]

An initial study of the potential of using ionizing radiation to control the growth of bacteria indicated that this approach could be useful in areas of industrial application. Accordingly, a program was started in 1967 on the possibility of using the radiation technique to control bacterial growth in industrial cutting fluids. These fluids often contain a hundredweight of bacteria per thousand gallons, causing chemical changes that significantly affect their engineering properties. Included in this experimental and theoretical program are an initial cost-benefit analysis of the method compared to other competitive procedures and the design, construction, and operation of an irradiation system. In the final phase of this program, the irradiator will be tested in an industrial plant, under actual operating conditions, to provide meaningful data for more detailed technical and economic appraisal.

EMULSION POLYMERIZATION OF VINYL ACETATE

Previous research work suggested that radiation-initiated emulsion polymerization of vinyl acetate could yield polymers of interest to industry, with very high molecular weight. An engineering study is in progress to study this process at a production level of several pounds per hour and to develop engineering experience that will be applicable to other emulsion polymerization systems.

Power Generators and Heat Sources

Use of the heat of radioactive decay as a direct source of thermal energy and for conversion to electric and mechanical power is one of the most rapidly advancing and broadly applicable of the current nuclear technologies. Technical feasibility of isotopically powered electric generators already has been amply demonstrated in space, terrestrial, and oceanographic applications. Proof-of-principle devices for direct use of heat and as sources of mechanical energy are currently under development. It is to be expected that complete families of isotopic heat sources and conversion devices will become available to serve a wide spectrum of requirements.

The first electric-power generators using isotopic heat were developed in the United States for the space program. These Systems for Nuclear Auxiliary Power (SNAP) have performed so well that the same principle has been applied for marine and terrestrial applications; now better systems are being developed for both space and terrestrial use. In addition to electric-power generation, radioisotope sources are also being studied for applications in which the heat is used directly.

Isotopic Fuels

The isotopes currently under development for space use are ^{238}Pu (half-life, 87.5 years) and ^{244}Cm (half-life, 18 years) for long missions and ^{210}Po (half-life, 138 days) for 90- to 120-day missions. Plutonium-238 and ^{210}Po are primarily alpha emitters, requiring little or no shielding to protect crews and payloads from radiation but requiring that provision be made to vent or contain the helium gas released by the fuel. The production of ^{210}Po is currently limited only by the facilities available for processing and encapsulating

large quantities. Production of ^{238}Pu is much more expensive and time-consuming, and large quantities require commitments of national resources several years ahead of the scheduled needs. Curium-244 has a higher power density than ^{238}Pu; however, the shielding required because of spontaneous neutron emissions may limit its utilization. Thulium-170, a beta emitter with a 130-day half-life, is also being considered for space missions of limited duration.

The USAEC is working now with small quantities of ^{244}Cm to learn about its physical and chemical properties. Curium can be produced from raw materials that are more readily available than the target material for ^{238}Pu and hence may assist in reducing potential future shortages of ^{238}Pu.

We are also investigating the use of ^{90}Sr for space. This beta-emitting isotope is much less expensive and more plentiful than the alpha emitters, and thus the fuel costs could be cut by a factor of about 10 compared to ^{238}Pu. Strontium-90 has the disadvantage of requiring considerably more shielding than any of the above alpha emitters, and its potential biological ingestion hazard is similar to that of the alpha-emitting fuels. Special remote-handling procedures would be required at the launch pad. For these reasons we are not yet sure of the use that will be made of ^{90}Sr fuels in space systems.

The ultimate use of any of these isotopes in the space program depends on finding chemical forms of the fuel and container materials which are compatible with each other and will operate for extended periods at 1500°C with ^{238}Pu and at 2000°C with ^{210}Po and ^{244}Cm.

For terrestrial applications, isotope system weight is less important than for space systems; shielding can be provided, thus allowing the use of fission products.

Strontium-90 has been used in most of the generators for terrestrial applications.[30] This nuclide is a fission product, and thus an abundant supply is available at a reasonable cost. Strontium titanate is the preferred fuel form at present. Strontium titanate is extremely stable and has very low solubility in both fresh and salt water. Hastelloy C is used for encapsulation. Certainly for the foreseeable future this isotope will continue to be preferred for terrestrial applications, and further work is being done to find even more suitable fuel forms.

Cerium-144 and ^{60}Co are both being studied as potential fuels in generator systems in the 1- to 10-kw(e) range. Cobalt-60, in particular, is beginning to assume considerable importance in the large heat-source program.

The Commission's Savannah River Plant[31] has recently produced ~10 Mc of ^{60}Co with an average specific activity between 300 and 400 curies/g. We have asked for proposals from industry for the design, fabrication, and testing of an experimental ^{60}Co-fueled heat-source demonstration unit that would operate at temperatures up to 850°C.

Space Systems

Radioisotope thermoelectric generators have been used successfully in two Navy experimental navigation satellites[32] (SNAP-3A, 3 watts*), which were launched in 1961 and are still operating, and in two Navy operational navigation satellites (SNAP-9A, 25 watts), which were launched in 1963 and 1964 and also are still operating. Systems now under development include the SNAP-19 and SNAP-27 radioisotope thermoelectric generators.

SNAP-19

This system is being developed to supplement the power of the NASA Nimbus-B weather satellite. Two 25-watt generators are to be mounted on the sensory ring of the spacecraft. An electrically heated system has been under test for several months, and two complete nuclear-fueled generators are now being tested for acceptance and qualification for a launch currently scheduled for early 1968.

SNAP-27

This generator is designed to supply 50 watts minimum power to the Apollo Lunar Surface Experiment Package for at least 1 year after it is left on the moon at the time of the first Apollo landing. The first qualification system is undergoing testing now, and the first flight generator was delivered this fall. The first heat source is due for completion in November of this year. This generator will be transported to the moon in a storage bin within the Apollo Lunar Excursion Module; the fuel is carried separately in its shipping container mounted outside the module. After landing, the astronauts assemble and fuel the generator. The fuel shipping cask is designed to maintain the fuel intact in case of an early mission abort, which could result in reentry. The unit now under test is producing over 70 watts and weighs about 41 lb; this low (0.6) weight to power ratio is achieved by extensive use of beryllium in the structural parts of the generator.

ADVANCED SYSTEMS

In addition to the systems being built for a specific mission, several others are under study for longer-range needs. In the SNAP-29 program we are developing the technology for isotopic generators at the 500-watt level for 90- to 120-day missions. Whereas the earlier units used ^{238}Pu fuel, the SNAP-29 generator, designed for shorter-lived missions, is able to use the cheaper and more readily available ^{210}Po fuel.

*Unless otherwise stated, "watts" refers to the electrical output of the generator.

The SNAP-29 generator unit is designed to adapt to various tasks, including long-life missions using ^{238}Pu fuel, and will weigh about 1 lb/watt. The SNAP-29 program is now concentrating primarily on fuel and component technology. If the results are promising, we will proceed toward our present goal of a fueled ground test in 1969, with flight tests possibly scheduled for 1970–1971.

We recently completed preliminary designs for a 250-watt generator. The feasibility of controlled intact reentry of the fuel into the atmosphere in the event of a launch or orbit injection malfunction has been investigated. This generator is of interest to several missions. During the coming year we plan to complete one of these designs and to build and test a mockup of the ground equipment needed for handling the large quantities of isotopes that would be required at the launch complex.

Terrestrial Systems

For several years the AEC has been actively pursuing the development of isotopic power systems for use in remote terrestrial and marine applications. This initial effort has largely concentrated on the design and development of the first generation SNAP-7 series of ^{90}Sr-fueled thermoelectric power systems.[33] Each of these systems is characterized by the use of lead telluride thermoelectric junctions operating at temperatures below 500°C. These relatively expensive proof-of-principle devices have been designed to demonstrate the feasibility of developing long-lived isotopic power systems that are capable of unattended and safe operation in a variety of hostile environments.

Important operating and test data have been obtained from these prototype systems, which have clearly demonstrated the capability of isotopic power sources for long-term unattended operation under severe environmental extremes without compromise of safety or device integrity. We are particularly proud of SNAP-7E, which has recently completed its third year of unattended operation at a depth of 15 thousand ft in the Atlantic Ocean.

The engineering data obtained from these prototypes have highlighted areas where significant technological advancements are required. Of immediate importance are improvements in thermoelectric material properties and stability; fuel characteristics and encapsulation; thermal management and insulation capability; materials properties and compatibility; and, finally, fabrication technology. These advancements in technology are being accomplished by an orderly and technically disciplined research and development program that will be discussed later. These programs should lead to the development of a second generation of isotopic power systems that possess the required reliability, operating lifetime, and economic advantages necessary to ensure widespread applicability.

The relative success of the SNAP-7 program has resulted in important changes in program philosophy.[34] We can now proceed with the development of isotopic power systems independent of any specific application. As a result of continuous assessment of potential needs, we have established meaningful design criteria and have initiated development within a number of discrete power levels. This approach should result in the attainment of a technological base that is adaptable to many potential applications. The necessity of initiating these programs prior to the establishment of firm mission requirements results from the long cycle associated with nuclear power development. Attempting to meet specific operational requirements with rigid time schedules does not lend itself to the orderly development of base technology. Our experiences repeatedly show that under these conditions the objective of establishing a sound base of technology is often compromised and long-term objectives sacrificed. As a result, the necessity to fully develop the base technology toward general areas of application and broadly defined needs for nuclear power is recognized. By working closely with interested user organizations, we can respond to changes in potential need and application.

MICROWATT ISOTOPIC POWER SYSTEMS

Following extensive technical discussions with medical research institutes, the USAEC initiated a program to develop a plutonium-fueled thermoelectric generator to power a surgically implantable cardiac pacemaker. While the effort is being concentrated on the pacemaker application, the basic engineering concept is expected to have important application to other biomedical stimulation problems.

Effort is currently concentrated on (1) minimizing the total necessary fuel inventory in order to reduce external radiation to medically acceptable levels; (2) developing efficient heat-rejection techniques; and (3) selecting electronic components of proven reliability to ensure attainment of the 10-year minimum operating lifetime. The basic phase of the program will be completed at the end of this fiscal year. Subsequent effort will involve an extensive fabrication and statistical test effort and will include animal in vivo testing.

100- to 1000-mw ISOTOPIC POWER SYSTEMS

A program is planned to design, develop, test, and demonstrate isotopic power systems in the 100- to 1000-mw range for specialized military and commercial applications, including oceanographic instrumentation, undersea cable boosters, and undersea weapons activation. Primary objectives are low cost, reliability, safety, compactness, and ease of handling. As

a first step, an applications engineering study was completed last year. From the data obtained, development programs will be initiated, concentrating on systems that offer the greatest probability for widespread application. The isotopic fuel selected will probably be ^{238}Pu or ^{90}Sr, depending on weight and cost considerations.

10- to 100-WATT ISOTOPIC POWER SYSTEMS

The major effort in this program is currently directed toward the development of a series of compact and highly reliable isotopic power systems in the 10- to 100-watt power range. This program should lead to the development of a second generation of radioisotope power systems that possess the required reliability, operating lifetime, and economic advantages necessary to ensure widespread applicability in marine and remote terrestrial environments. This important program consists of two specific projects: (1) SNAP-21, which is a two-phased effort to develop ^{90}Sr-powered 10- and 20-watt generators for deep-sea applications; and (2) SNAP-23, designed to develop and fabricate ^{90}Sr-powered generators having capacities of 25, 60, and 100 watts for remote terrestrial applications.

KILOWATT POWER SYSTEMS

The USAEC has become increasingly aware of the potential needs for large isotopic plants—for man-in-the-sea type activities, for offshore oil exploration, and for manned and unmanned research stations. Our evaluation and assessment of these potential needs have resulted in initiation of a research and development program directed toward the eventual production of isotopic powered systems in the 1- to 10-kw range. The first phase of this program is an extensive applications engineering and design study to evaluate alternative energy-conversion cycles, plant configurations, isotope fuel requirements, performance requirements, environmental constraints, and economic criteria. The isotopes of interest in this program are ^{90}Sr, ^{60}Co, and ^{144}Ce.

TECHNICAL PHILOSOPHY

Each of the programs described is characterized by a systematic and orderly engineering approach established primarily to assure attainment of technical objectives. Such a disciplined approach is necessary to meet the stringent performance objectives, including unattended operating lifetimes of 5 to 10 years. A premium is therefore being placed upon use of rigid quality assurance and reliability techniques, together with rigorous and long-term proof testing of all materials and components under laboratory and simulated environmental conditions. It is expected that such quality assurance and testing activities will be successful in identifying design problems associated with long-term high-temperature operating prior to delivery and installation of prototype systems. Furthermore, this approach, with its emphasis upon engineering details, quality assurance techniques, and proof testing, will assure that the technology incorporated into the systems is of a known and predictable state.

To obtain long-term performance data under real environmental conditions, cooperative test programs are being developed in conjunction with prospective user organizations. Fully instrumented prototypes will be installed under varying environmental conditions for purposes of providing meaningful and timely data necessary to evaluate internal thermal stability; heat-transfer characteristics associated with marine fouling and ocean-bottom burial; electrical performance of major subcomponents; and long-term materials behavior. The need for extensive test data of this type is essential to a full understanding of device behavior.

Radioisotope Power System for Artificial Heart

The National Heart Institute has carried out a study to determine the need for a circulatory support system, to identify key technological problems requiring solution, and to forecast the time and money required for the development of such devices. This study indicated that heart disease claimed about 700,000 American lives this year and that about 100,000 of these might have been helped if a circulatory support system were available.

The National Heart Institute has undertaken a program to develop a series of devices beginning with a temporary assist device such as the DeBakey left ventricle assist pump and ending with a totally implantable system to replace the biological heart.

The AEC has selected four firms to conduct parallel conceptual design studies of a radioisotope-powered engine to actuate blood pumps that would assist or replace a diseased or damaged heart. The heat from the decay of the radioisotope would be used by a suitable converter to provide power for the artificial heart. The encapsulated heat source would likely be ^{238}Pu or ^{147}Pm. In addition to the heat source, the engine would include a thermodynamic converter, a heat exchanger using blood as the cooling medium, and a control system to regulate the power output between 1 and 7 watts. Details of this program are discussed in a paper by W. H. Pennington to be presented at this conference.

Isotopes Production and Sales

Over the past 5 years, significant changes have taken place in the United States in methods used for the production, distribution, and sale of radioisotopes. Many of these have been described at previous Japan Conferences on Radioisotopes.[35–37] I will summarize

the effects these changes have had and the current situation, as well as identify new developments.

Sales of Isotopes and Services by the USAEC

Although the production and sale of many isotopes have been assumed by private industry, USAEC contractors still supply a large number of these products. The major sales by Commission facilities in terms of quantity are for kilocurie amounts of separated fission products and ^{60}Co (in quantities of 100,000 curies or more and with a specific activity up to 45 curies/g) or for service irradiations in reactors or cyclotrons. These sales for the past 3 years are summarized in Table 3 and Fig. 2; sales of six isotopes—^{241}Am, ^{14}C, ^{60}Co, ^{137}Cs, ^{3}H, and ^{90}Sr—accounted for well over half the total value of isotope sales (Table 4).

FISSION PRODUCTS

Last year at this meeting,[35] it was mentioned that a private industrial firm, Isochem Inc., was planning to build a facility for the large-scale separation and encapsulation of the fission products ^{137}Cs, ^{90}Sr, and ^{147}Pm. Plans for this plant have now been terminated. The USAEC facilities will therefore be used to continue production and sales of these materials. Currently 5.6 Mc of ^{90}Sr is available for sale or is being processed at ORNL, and Hanford has about 1 Mc separated and awaiting shipment to ORNL. Oak Ridge National Laboratory also has about 1.5 Mc of ^{137}Cs in inventory available for sale.

Table 3 SALES OF ISOTOPES AND SERVICES BY THE USAEC[38]

	Commercial sales	USAEC project sales	Total
	Fiscal Year 1965		
Isotopes	$1,939,950	$ 864,463	$2,804,413
Cyclotron irradiation	246,094	12,571	258,665
Reactor irradiation	78,879	99,270	178,149
Special services	108,346	183,525	291,871
Total	$2,373,269	$1,159,829	$3,533,098
	Fiscal Year 1966		
Isotopes	$2,344,721	$1,416,939	$3,761,660
Cyclotron irradiation	263,682	14,031	277,713
Reactor irradiation	146,971	68,743	215,714
Special services	118,484	45,626	164,110
Total	$2,873,858	$1,545,339	$4,419,197
	Fiscal Year 1967		
Isotopes	$1,838,059	$1,436,663	$3,274,722
Cyclotron irradiation	240,838	17,398	258,236
Reactor irradiation	264,687	121,247	385,934
Special services	125,338	72,838	198,176
Total	$2,468,922	$1,648,146	$4,117,068

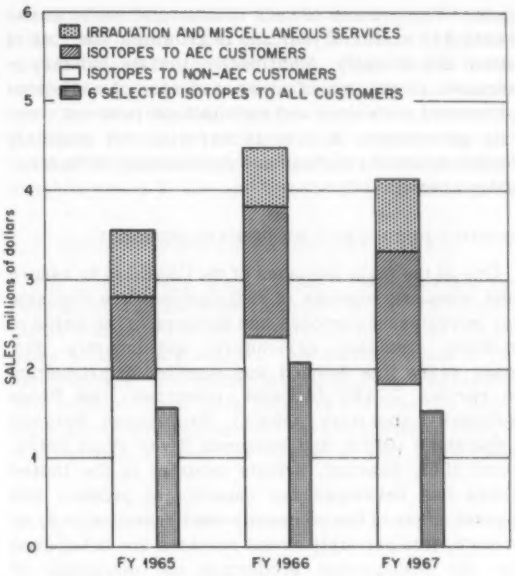

Fig. 2 Sales of radioisotopes and services by the USAEC.

Sales of Isotopes and Services by Private Industry

The radioisotope and radiation industry in the United States is characterized by significant participation of private companies. Private enterprise is involved in major aspects of radioisotope distribution, including private production of radioisotopes and commercial sale of radiochemicals, radiopharmaceuticals, radiation sources, and nuclear devices such as

Table 4 SALES OF SIX ISOTOPES BY THE USAEC

Isotope	Fiscal year 1965 Activity, curies*	Fiscal year 1965 Value	Fiscal year 1966 Activity, curies*	Fiscal year 1966 Value	Fiscal year 1967 Activity, curies*	Fiscal year 1967 Value
^{241}Am	546 g	$ 657,646	1101 g	$ 745,192	421 g	$ 481,636
^{14}C	64.6	261,925	158	320,684	52	199,913
^{137}Cs	350,986	210,893	70,002	27,107	567,986	86,363
^{60}Co	1,297,464	262,153	1,311,060	594,986	2,327,117	421,937
^{3}H	120,571	130,601	826,981	274,924	672,217	243,859
^{90}Sr	1,015	3,570	109,262	38,992	396,282	82,492
Total		$1,526,788		$2,001,885		$1,516,200

*The ^{241}Am values are in grams, not curies.

gages. The volume of such commercial business exceeds $40 million a year and is growing at the rate of about 20% annually. Additionally, private industry is engaged in the manufacture and sale of radiation-processed materials and radioisotope-powered electric generators. It also is carrying out privately funded selected research and development in the foregoing areas.

ISOTOPE PRODUCTION BY PRIVATE INDUSTRY

One of the main missions of the USAEC is to assure that adequate supplies of radioisotopes are available for government purposes and to users in the fields of science, medicine, agriculture, and industry. For many years this demand was supplied by production at various USAEC facilities, principally Oak Ridge National Laboratory (ORNL), Brookhaven National Laboratory (BNL), and Savannah River Plant (SRP). Since 1961, however, private industry in the United States has developed the capacity to produce and market many of the commonly used radioisotopes. At present, five privately owned reactors are being used for the commercial production and marketing of radioisotopes. The USAEC, in accordance with stated public policy, has withdrawn from production of these isotopes as commercial capability has become available. To date the USAEC has withdrawn from the routine production of 37 radioisotopes; these are listed in Table 5.

Table 5 RADIOISOTOPES FROM WHICH USAEC HAS WITHDRAWN

Antimony-124	Iron-55
Antimony-125	Iron-59
Arsenic-76	Lanthanum-140
Arsenic-77	Mercury-197
Bromine-82	Mercury-203
Cadmium-109	Molybdenum-99
Cadmium-115	Molybdenum-99–technetium-99*m* generator
Cadmium-115*m*	Phosphorus-32
Calcium-45	Potassium-42
Cerium-141	Selenium-75
Cesium-134	Silver-110
Chromium-51	Sodium-24
Cobalt-58	Strontium-85
Cobalt-60 (metallic sources)	Strontium-87*m*
Copper-64	Sulfur-35
Gold-198	Tin-113
Gold-199	Zinc-65
Iodine-125	
Iodine-131	
Iridium-192 (metallic sources)	

These withdrawals have been made primarily during the past 2 years. Prior to each withdrawal, the USAEC makes a determination that an adequate, continuing supply of these radioisotopes would be available from private sources at a reasonable price.

In 1961, both the Westinghouse Electric Corporation and General Electric Company started producing limited quantities of ^{60}Co in their test reactors to serve the medical teletherapy and industrial radiography markets. The Westinghouse reactor was subsequently shut down, and until recently the General Electric Company was the only commercial firm in the United States producing substantial quantities of ^{60}Co. In 1966, Neutron Products, Inc., started a joint program with Consumers Power Company of Jackson, Mich., at its Big Rock Point Nuclear Power Station to produce megacurie quantities of ^{60}Co, having a specific activity of 15 to 80 curies/g. This is the first commercial production of a radioisotope in a U. S. power reactor. Neutron Products, Inc., has recently announced prices and current availability of the ^{60}Co.

RADIOCHEMICALS

About 19 privately owned firms are engaged in the commercial distribution of radiochemical materials. The industry sales volume is approximately $8 million annually and is growing at the rate of 15% a year.

RADIOPHARMACEUTICALS

Annual radiopharmaceutical sales total $12 million and are growing by 20% a year. This reflects the widespread use of radioisotopes for medical purposes. Each year between 3 and 4 million patients are examined with radioisotopes. This market is served by eight private firms who between them distribute annually approximately 20,000 curies of radiomaterials in about 350,000 shipments.

RADIATION SOURCES

Radioisotopes for use as radiation sources for process radiation applications, medical teletherapy, industrial radiography and process control, and calibration sources, together with associated equipment and facilities, are available from 18 private companies. The radiation source business totals about $3 million annually and is increasing by 20% a year. Additionally, eight firms supply electron accelerators for processing purposes. The radiation source business has potential for rapid and dramatic growth, reflecting the great potential of process radiation applications and radiation preservation of foods.

GAGING

Radioisotope gaging has achieved widespread acceptance in U. S. industry. Approximately 5000 such devices currently are in routine use. The market is served by nine private firms with total sales of about $20 million annually. The market is expanding by 15% a year.

RADIATION-PROCESSED MATERIALS

The use of radiation by private industry as an industrial processing tool has been steadily increasing in the United States. The value of materials processed

in the United States by radiation was estimated to be $250 million in 1966. The installed process radiation capacity, estimated as 275 kw in 1965, had reached about 400 kw in 1966. This growth rate of 30% per year is expected to continue or increase over the next several years. Over 90% of this installed process radiation capacity currently is from electron-beam irradiators. In the past few years, however, several commercial irradiation facilities using radioisotopes have been completed. Over the past year, particularly, there has been increasing evidence that several new processes developed by industry will use radioisotopes as radiation sources for commercial-scale operations. Almost two-thirds of this installed radiation capacity is used to process plastics and manufacture chemicals; the remainder is almost equally divided among sterilization of hospital supplies, manufacture of semiconductors, modification of textiles, and miscellaneous processes.

Several new industrial processes using irradiation have been publicized in the past year. Probably the most important of these, from the standpoint of economic potential, were two systems that were announced for application of coatings to both wood and metals. The Ford Motor Company announced their Electrocure process, and Radiation Dynamics, their Dynacote process.

Ford is planning to use the Electrocure process as a substitute for painting several metal parts in automobile manufacture. Because of the high processing volume in the automobile industry, this could amount to a sizable increase in the use of radiation. The process has also been licensed for coating wood to Boise Cascade, who is building a facility with a capacity for handling 10 million sq ft/year of exterior prefinished pigmented siding.

Workers at Radiation Dynamics have recently analyzed the economics of their Dynacote process compared with commercial painting and heat drying.[39] The comparison showed that the raw materials for the Dynacote process cost about one-half as much as those for a paint process—the capital costs being about two-thirds and the operating costs about one-third of those of the conventional process.

Using radiation to apply a durable-press finish to fabrics has recently been announced as a commercial process by two companies. Although few details of these processes are available, it is known that they use an electron accelerator as a source of radiation. One of these processes also produces a fabric from which stains can be more easily removed than from other fabrics.

RADIOISOTOPE-POWERED GENERATORS

Radioisotope power sources were originally developed for the U. S. space program. Because of the advantages of these generators, such as light weight, portability, long life, and lack of dependence on other forms of energy, such as the sun in the case of solar cells, they were also adapted for terrestrial and marine uses. In the past year, three firms in the United States and one in Great Britain have announced that they are offering these devices for sale as off-the-shelf items. In addition, one company has announced it has developed, fabricated, and qualified isotope heaters of 2, 2.5, and 5 watts (thermal).

U. S. ISOTOPE PRODUCTION AND SALES IN TOTAL

The U. S. Bureau of the Census[40] estimated that the volume of radioisotopes shipped from non-USAEC plants had grown to $5.6 million in 1966. The USAEC sales totaled $4.4 million. Commercial sales of radiochemicals and radiopharmaceuticals aggregated $20 million. Thus U. S. sales of radioisotope materials alone have reached the very respectable total of $30 million a year, as shown in Fig. 3. To these

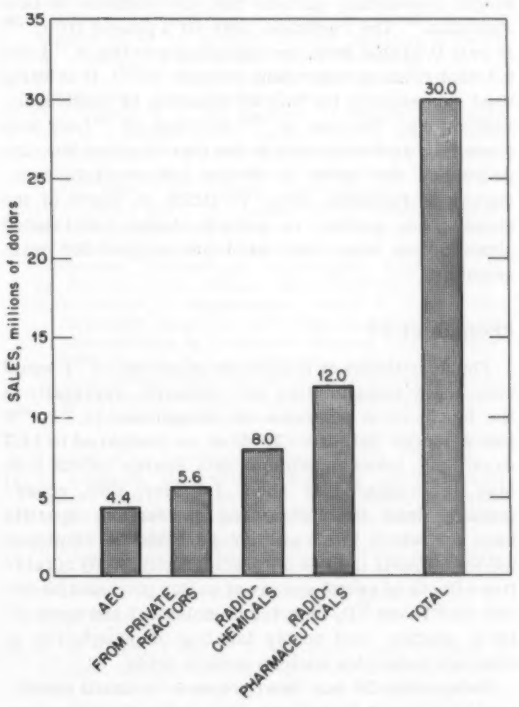

Fig. 3 U. S. radioisotope sales in 1966.

sales must be added the sales of radioisotope devices, such as gages, and of radiation-processed materials. Looked at from this overall viewpoint, it can be seen that radioisotopes are generating commercial activity in the United States at a level of several hundred million dollars annually.

Development of New Isotopes by the USAEC

As private industry has assumed increasing responsibility for routine production of many isotopes, research workers at USAEC radioisotope production sites have been able to concentrate more on the development of methods for producing new isotopes having importance in many fields of research, particularly in medicine. Examples of such radioisotopes are ^{123}I, ^{33}P, ^{69m}Zn, ^{83}Rb, and ^{67}Cu. Several new devices for extracting short-lived daughters from longer-lived parents (the commonly termed "radioisotope cows") have also been developed.

Iodine-123

Iodine-123 (13 hr, 0.159 Mev gamma) closely approaches the ideal radionuclide for use in medical diagnostic procedures because of its short half-life, single low-energy gamma ray, and absence of beta radiation.[41] The radiation dose to a patient from ^{123}I is only 0.03 that from an equivalent quantity of ^{131}I and 0.1 that from an equivalent quantity of ^{125}I. It is being used successfully for thyroid scanning by medical investigators. The use of ^{123}I in place of ^{131}I not only gives less radiation dose to the thyroid gland but also decreases the error in thyroid iodine-uptake measurement resulting from variation of depth of the gland from patient to patient. Iodine-123-labeled albumin has also been used as an agent for brain scanning.

Phosphorus-33

The availability of millicurie quantities of ^{33}P would open many opportunities for research, especially in the biochemical sciences. In comparison to ^{32}P, ^{33}P has a longer half-life (25 days as compared to 14.3 days) and lower maximum beta energy (about 0.25 Mev as compared to about 1.7 Mev) with, consequently, less bremsstrahlung production. Specific uses for which this radioisotope could be employed advantageously include autoradiography, study of relative effects of recoil energy of sulfur produced by decay of ^{32}P and ^{33}P, long-term ecological and agricultural studies, and double labeling of phosphorus in complex molecules such as nucleic acids.

Phosphorus-33 has been prepared in small quantities by several techniques, but yields have been too low for production of millicurie amounts because of lack of suitable targets. Workers at ORNL have recently produced millicurie amounts of this isotope by fast-neutron irradiation[42] of sulfur targets enriched in ^{33}S or potassium chloride targets enriched in ^{36}Cl followed by aging to allow decay of the ^{32}P. Irradiation of sulfur targets enriched in ^{33}S with a thermalized neutron flux has also been successful.[43] Current work is concerned with preparation and evaluation of targets enriched to about 90% ^{33}S. With such targets it will be possible to prepare ^{33}P containing only 5 to 10% ^{32}P; this material will require much less time for the ^{32}P to decay and thus yield higher-purity ^{33}P.

Zinc-69m

Zinc-69m (14 hr, 0.44 Mev gamma) has recently been used to demonstrate uptake of zinc by the prostate in humans.[44] The ^{69m}Zn is prepared by neutron irradiation of 99.3% ^{68}Zn. This experiment followed similar tests in animals[45] with ^{65}Zn that was not suitable for human use because of its radiation properties (243 days, 1.1 Mev gamma).

Indium-113m

Indium-113m (1.7 days, 390 kev gamma) "milking" devices are now available commercially from several companies in the United States. The parent isotope in these devices is ^{114}Sn. The ^{113m}In has been used medically for scanning many organs[46–49] in humans, such as the lung, brain, and blood pool, and some believe that it may replace ^{99m}Tc in most medical uses because the half-life of the parent ^{113}Sn (118 days) is longer than that of the ^{99m}Tc parent, ^{99}Mo (66.7 hr).

An inexpensive radioisotope milking device for classroom demonstration was developed by ORNL using the ^{137}Cs (half-life, 30 years) to ^{137m}Ba (half-life, 2.6 min) system.[50] Oak Ridge National Laboratory has supplied several of these devices to physicians for evaluation in diagnostic procedures. One of these devices is being used by Dr. Masahiro Iio at the University of Tokyo.

High Flux Reactors

Within the last year the High-Flux Isotope Reactor (HFIR)[51] at ORNL has been brought to full power and operated routinely. Designed and built for the production of transplutonium isotopes, the reactor has a power density of 1 Mw per liter of core volume, a thermal flux of about 1×10^{15} n/(cm^2)(sec) and a fast flux of about 1×10^{15} n/(cm^2)(sec). The high thermal flux will make the use of double neutron-capture reactions possible, such as $^{64}Ni(2n,\gamma)^{66}Ni$, while the fast flux will make possible (n,p) reactions, such as $^{54}Fe(n,p)^{54}Mn$, as well as (n,α) and $(n,2n)$ reactions that were not feasible at lower fluxes.

The Savannah River Plant[52] modified one of its production reactors and operated it for about a year at a thermal flux of 1×10^{15} n/(cm^2)(sec). With this high flux the reactor was used in the transplutonium-element production to advance the program about a year by preirradiation of HFIR target material. A sample of ^{60}Co having a specific activity of 700 curies/g as well as small amounts of ^{244}Cm and ^{252}Cf were also produced, along with ~10 Mc of 300–400 curies per gram of ^{60}Co. A portion of the

latter material will be used in a long-term, 30 kw(t), ^{60}Co heat-source experimental program.

Recently workers at ORNL made 20 g of ^{242}Cm (half-life, 162 days). I view this as a truly outstanding technical achievement, particularly when one considers that previously not more than milligram quantities had been produced and separated. This material was prepared by irradiation of about 55 g of ^{241}Am in the Oak Ridge Research Reactor and then by separation of the resulting ^{242}Cm in the pilot plant for the Transuranium Facility. The ^{242}Cm oxide was processed into pellets, each of which produced 200 watts(t), and 7.5 g (25 kilocuries) were used to power a 20-watt electrical generator designed for use on a spacecraft. The device operated for 90 days under simulated conditions of lunar day and night. The lunar day–lunar night temperatures used in the experiment ranged from 235 to −235°F. The intense alpha heating generated by this source makes it the most concentrated isotopic power source ever produced in the world to our knowledge—120 watts(t)/g.

Private Isotope Technology Research and Development

As is readily understandable, information on privately sponsored research and development programs in isotopes and radiation technology is not generally available. The advent of new radioisotope products, new radiation processes, and new radioisotope devices all give evidence, however, of continuing and significant privately funded effort. It is particularly noteworthy, for example, that radiopharmaceutical manufacturers are beginning to do their own original research as opposed to their earlier reliance on the efforts of individual investigators in universities and medical institutions. The development and marketing of ^{113m}In "cows"[48,49] for medical use, for example, are indicative of such original effort. Similarly the development of free-air backscatter thickness gages and new applications in the measurement of materials applied as a coating represent the fruits of privately sponsored research and development. It is not possible to determine precisely the dollar magnitude of privately funded effort in developing isotopes and radiation technology. It appears, however, that this exceeds $20 million annually.

Acknowledgments

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(Robert H. Lafferty, Jr.)

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Section I

Isotopes and Radiation Technology

Isotope Production and Development

Cobalt-60 for Power Sources*

By C. P. Ross†

Abstract: The status of the program at Savannah River Laboratory for the development of ^{60}Co as a radioisotopic power source is reviewed. This paper, which discusses interest in and use of ^{60}Co as a power source, includes coverage of applications, properties, shielding and containment, production and cost, and capsule material selection, fabrication, and testing.

Cobalt-60, one of the radioisotopes being considered for large-scale isotopic power sources on land, under sea, and in space, is now available in quantities and at activities suitable for demonstrations.

With a power density and physical properties that are suitable for many applications, with large-quantity production at low cost (hundreds of megacuries per year at <25 cents per curie), and with a half-life of 5.24 years, ^{60}Co provides a convenient compromise among power density, production, utilization, and disposal. Radioisotopic power sources of ^{60}Co may compete favorably with the small SNAP reactors for space applications and with the larger sized ^{90}Sr land-based radioisotopic power sources.

Under the sponsorship of the U. S. Atomic Energy Commission (AEC), the Savannah River Laboratory (SRL) is engaged in developing technology for efficiently producing ^{60}Co in large quantities, in selecting suitable encapsulating materials for high-activity cobalt, and in determining long-term operating limits of capsules.[1,2]

Applications

Isotopic heat sources have been proposed for use in many applications where they may have advantages over other power supplies,[3] e.g., manned stations such as Sealab and Antarctic experimental and weather stations, and unattended weather stations, navigation aids, communication facilities, and surveillance devices. Studies have suggested that ^{60}Co could replace nuclear reactors in certain space applications.[4,5] Isotopic power sources such as ^{60}Co would provide a simpler system, but reactors would have the potential for lower fuel cost and would be safer on the launch pad and on reentry if they can be shut down well ahead of time. Isotopic power sources may need a mechanism to dump excess thermal power at beginning-of-life and when electric-power requirements are reduced, whereas reactors need a system for startup and control of thermal power.

The Naval Facilities Engineering Command has surveyed[6] future possible Navy needs for remote power supplies and has an impressive list of requirements for power supplies of 1 to 10 kw(e) as well as of 10 to 100 watts(e).

The Division of Isotopes Development of the AEC has initiated a program that is aimed at demonstrating the integrity of ^{60}Co heat sources that can produce useful energy for long times. The program should provide basic data that can be used to develop heat sources suitable for integration with conversion equipment. Proposals from several companies are being evaluated.[7]

The Division of Reactor Development and Technology has started a program[8] that will evaluate ^{60}Co and the fission products ^{90}Sr and ^{144}Ce as to their applicability for power supplies from 5 to 100 kw(t). This study will first compare the limiting characteristics of the individual heat sources and then compare the limiting characteristics of the heat sources when coupled with various power-conversion systems. Figure 1 compares the specific power of fuel forms of these isotopes as a function of decay time.

Properties

Irradiation of natural cobalt (^{59}Co) in a nuclear reactor produces ^{60}Co, which decays by emitting beta particles and gamma rays to form stable ^{60}Ni (Fig. 2).

*The information contained in this article was developed during the course of work under Contract AT(07-2)-1 with the U. S. Atomic Energy Commission.

†Savannah River Laboratory, E. I. du Pont de Nemours & Co., Inc., Aiken, S. C. 29801.

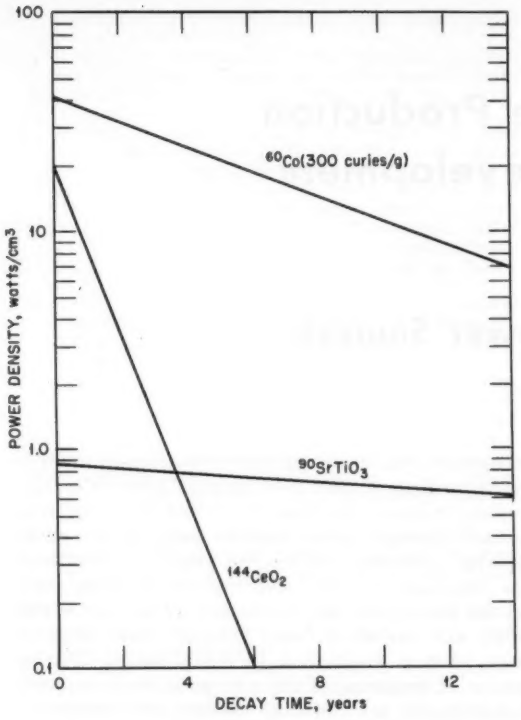


Fig. 1 Power density of radioisotope fuels.

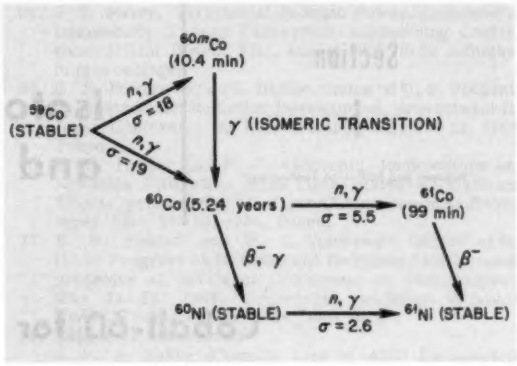


Fig. 2 Production process for ^{60}Co.

Nickel has similar properties and forms a solid solution with cobalt over the full range of composition. Hence the properties of natural cobalt have generally been used in assessing the characteristics of ^{60}Co heat sources. However, measurements have been made at SRL to determine the chemical composition, dimensions, density, hardness, melting point, and microstructure of irradiated cobalt metal.[9]

Metallographic examinations revealed no significant dimensional or structural changes in cobalt metal irradiated to 700 curies/g (96 watts/cm³, the record activity) or in cobalt metal irradiated to 300 curies/g and allowed to decay for more than one half-life. The only significant property change observed was an increase in hardness due to irradiation and to the buildup of nickel as the ^{60}Co decayed (Table 1). This increase should not seriously affect the properties of a heat source of cobalt capsules. These measurements were made on cobalt wafers that had been obtained commercially, plated with ½ to 1 mil of nickel to minimize the spread of contamination after irradiation, and irradiated at Savannah River. (The typical composition and future specifications of the cobalt raw material are given in Table 2.)

Other important physical properties of cobalt metal, related to its use as a high-temperature heat source, are given in Table 3.

The melting point of cobalt metal is 1495°C, and addition of up to 25% nickel will decrease the melting point only about 10°C. This melting point makes ^{60}Co applicable for all currently proposed power-conversion systems except thermionics,* and it may be possible to fabricate a fuel form of ^{60}Co that is stable at the higher temperatures required by thermionics.[10]

*In U. S. Patent 3,306,045 issued Feb. 28, 1967, W. H. Buford, Jr., and A. N. Thomas, Jr., of the Marquardt Corp., Van Nuys, Calif., proposed a system in which heat from a thermally shielded ^{60}Co source is supplied to a refractory metal at a much higher temperature.

Table 1 EFFECT OF IRRADIATION ON DENSITY AND HARDNESS OF COBALT

Specific activity, curies/g		Calculated composition at time of measurements, %			Density, g/cm³	Average hardness, R_D scale
At reactor discharge	At time of measurements	^{59}Co	^{60}Co	Ni		
Unirradiated		100	-	-	8.85	15
6	~6	~100	<1	<0.1	8.82	36
93	~93	91	8	1	8.85	36
100	50*	91	4	5	8.72	45
300	140	65	13	22	8.77	49
700	~700	27	62	11	8.93	43

*Pellets were 1/16 in. diameter by 1/16 in. high; all other samples were wafers 0.75 in. diameter by 0.040 in. thick.

Table 2 CHEMICAL COMPOSITION OF COBALT RAW MATERIAL

Element	Tentative specification Minimum wt.%	Tentative specification Maximum ppm (wt.)	Typical current analysis wt.%	Typical current analysis ppm (wt.)
Co + Ni	99.9		99.3	
Ni		1500		3800
Fe		1000		140
Cu		100		15
O		100		300
Si		100		3000
Th		100		<1
U		100		<1
Al		50		140
Cd		50		<1
Mn		50		3000
Pb		50		1
S		50		15
Cr		20		30
Mo		20		<1
V		20		1
W		20		6
P		10		5
B		5		1
Gd		5		<1
Li		5		<1

Table 3 SUMMARY OF PROPERTIES OF COBALT METAL
(Data from Ref. 11 Except As Noted)

Melting point	1495°C Impurities, including nickel decay product, decrease melting point by <20°C	
Vapor pressure	Temp., °C	Pressure, atm
	1050	7.1×10^{-9}
	1200	2.9×10^{-7}
	1595	9.6×10^{-6}
	2027	1.0×10^{-3}
	2327	1.0×10^{-2}
	3097	1.0
Specific heat	Temp., °C	Specific heat, cal/(g)(°C)
	0	0.099
	100	0.106
	500	0.126
	1100	0.204
	1400	0.168
	Molten	0.141
Thermal conductivity[12]	Temp., °C	Conductivity, cal/(sec)(cm)(°C)
	50	0.22
	100	0.20
	150	0.19
	500	0.13*
	1000	0.09*
Coefficient of thermal expansion[13]	14.2 μin./(in.)(°C) between 20 and 300°C 15.7 μin./(in.)(°C) at 400°C 16.8 μin./(in.)(°C) at 750°C	
Density	8.8 g/cm³ (solid forms)	
Oxidation rate in air[14]	Temp., °C	Parabolic scaling constant, $(mg/cm^2)^2/hr$
	400	0.0004
	600	0.1
	800	2.1
	1000	70.0
	1200	400.0

*Calculated from electrical resistivity.

Shielding and Containment Required

In decaying to ^{60}Ni, ^{60}Co emits beta particles with a maximum energy of 0.312 Mev and gamma rays with energies of 1.172 and 1.333 Mev. Since only a small fraction of the energy is absorbed in the cobalt [depending on the thickness as shown in Fig. 3 (Ref. 15)], a susceptor of some heavy metal must surround the cobalt to recover the energy. About 2 in. of tungsten or uranium shielding will recover over 99% of the total energy available, which is 1.56 watts/100 curies. Small amounts of other radiations are also emitted (Fig. 4), but these have negligible influence on shielding requirements or heat generation. The weight of shielding can be reduced by using cobalt with a high specific activity to reduce the size of the source. The shield weight for a heat source operating in a manned environment may be as little as 1000 lb for a 2-kw(t) source and 2000 lb for a 20-kw(t) source.[16]

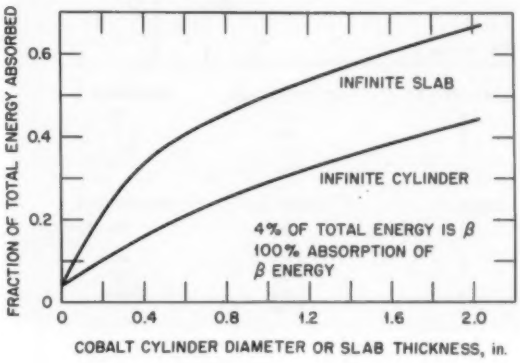


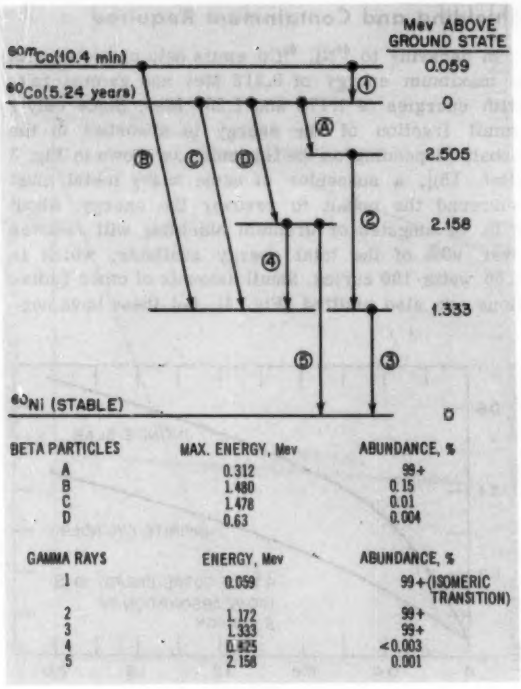
Fig. 3 Self-absorption of cobalt metal shapes.

When cobalt wafers are stacked in assembled capsules, the internal heat generation is increased and the heat-transfer area per wafer is decreased. Consequently the surface heat flux of the capsules increases and can present problems in handling. Calculated temperatures on the surface of capsules in air are shown in Fig. 5.

The biological hazard of ingestion of ^{60}Co is less than that for most of the isotopes being considered for heat-source application (Fig. 6). The relative hazards shown are based on the maximum permissible intake (MPI) over a short period that results in a limiting dose to the critical organ.[17]

Production and Cost of ^{60}Co

Cobalt-60 has been produced for many years at relatively low specific activity for use as radiation sources in research, teletherapy, and food irradia-



BETA PARTICLES	MAX. ENERGY, Mev	ABUNDANCE, %
A	0.312	99+
B	1.480	0.15
C	1.478	0.01
D	0.63	0.004

GAMMA RAYS	ENERGY, Mev	ABUNDANCE, %
1	0.059	99+(ISOMERIC TRANSITION)
2	1.172	99+
3	1.333	99+
4	0.825	<0.003
5	2.158	0.001

Fig. 4 Decay of ^{60}Co.

tion. Heat and power sources, however, require high specific activity cobalt. During the last 3 years, high-flux operation of the heavy-water-moderated Savannah River Plant reactors has made practical the production of large quantities of high-activity cobalt in a short time.[18]

Cobalt-60 can be produced either as the major product of a reactor charge or as one of several products in mixed-lattice charges. Cobalt metal must be irradiated in relatively thin sections (slabs or wafers less than $\sim 1/8$ in. thick, rods or pellets less than $\sim 1/4$ in. in diameter) for uniform activation and to minimize flux depressions. Sizes and shapes irradiated to date have had maximum widths or diameters of <1 in.; it is possible to irradiate a variety of shapes in target positions that are up to $3\frac{1}{2}$ in. in diameter.

Nearly 9 Mc of high-activity cobalt have been produced at Savannah River for development and demonstration of heat sources (Table 4). This quantity includes over 5 Mc as 0.745-in.-diameter wafers that have specific activities between 300 and 400 curies/g. The Division of Isotopes Development plans to use 2 Mc of this material for its demonstration heat-source program (see above).

The cost of producing ^{60}Co depends upon the specific activity desired. The activity of pure ^{60}Co is 1140 curies/g; however, the maximum activity of radioactive cobalt metal will be much lower because of burnup and decay. Large quantities of ^{60}Co cannot be produced economically above about 450 curies/g.

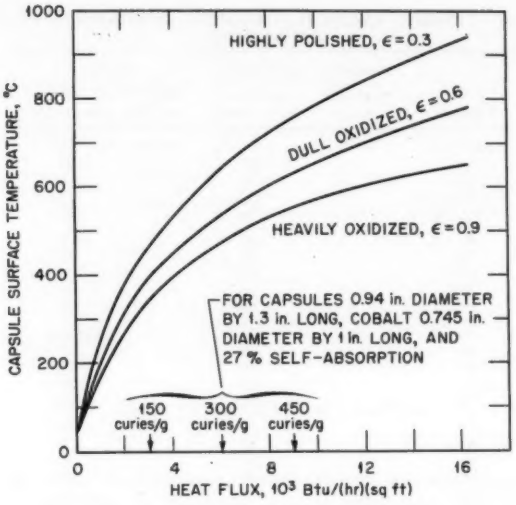


Fig. 5 Capsule surface temperature with natural convection and radiation to air.

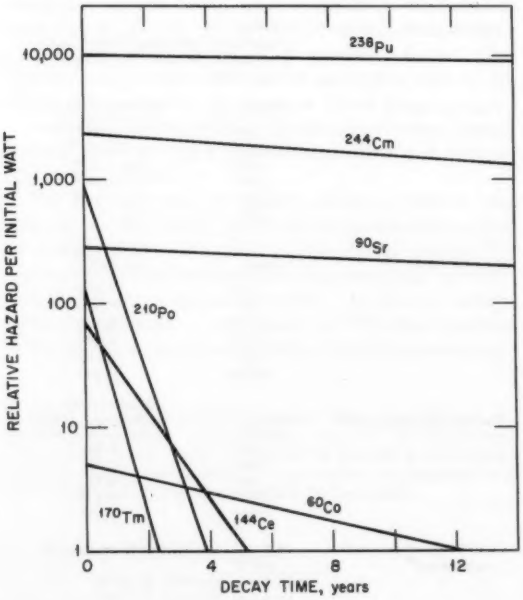


Fig. 6 Relative ingestion hazard for dispersed isotopes. (Short-period exposure.)[17]

Table 4 COBALT-60 FOR HEAT SOURCES*

Cobalt shape	Average activity, curies/g	No. of pieces	Total activity, Mc	Total power, kw
0.745-in. wafers				
Fuel form: cobalt metal (wrought)	400	2280	2.27	35.4
Fuel shape: wafers 0.745 ± 0.001 in. in diameter and	350	912	0.80	12.5
0.040 ± 0.003 in. thick,	330	1520	1.25	19.5
including 0.0005 to 0.001-in. nickel plate	300	1520	1.14	17.8
Cobalt density: 8.80 ± 0.05 g/cm³	320	152	0.12	1.9†
		6384	5.58	87.1
0.800-in. wafers				
Fuel form: cobalt metal (sintered)	390	255	0.28	4.4
Fuel shape: wafers 0.800 ± 0.001 in. in diameter and	410	136	0.16	2.5‡
0.040 ± 0.003 in. thick,	350	136	0.13	2.0§
including 0.0005 to 0.001-in. nickel plate		527	0.57	8.9
Cobalt density: 8.60 ± 0.10 g/cm³				
Nickel-plated slabs				
Fuel form: cobalt metal (wrought)	390	45	0.29	4.5
Fuel shape: slabs 3.00 ± 0.03 in. long,	360	48	0.29	4.5
0.640 ± 0.002 in. wide, and		93	0.58	9.0
0.060 ± 0.001 in. thick,				
including 0.0005 to 0.001-in. nickel plate				
Cobalt density: 8.80 ± 0.05 g/cm³				
Stainless-steel-canned slabs				
Fuel form: cobalt metal (wrought)	390	45	0.24	3.7
Fuel shape: slabs 2.96 ± 0.03 in. long,	330	48	0.22	3.4
0.735 to 0.740 in. wide,		93	0.46	7.1
0.092 ± 0.001 in. thick,				
sheath thickness 0.015 in. minimum, and				
cobalt dimensions same as nickel-plated				
slabs above except 2.44 in. long				
Cobalt density: 8.80 ± 0.05 g/cm³				
Stainless-steel-coextruded slabs				
Fuel form: cobalt metal (wrought)	390	60	0.27	4.2
Fuel shape: slabs 3.00 ± 0.03 in. long,	360	64	0.27	4.2
0.740 ± 0.002 in. wide,		124	0.54	8.4
0.072 ± 0.002 in. thick, and stainless-				
steel thickness 0.015 in. minimum				
cobalt dimensions: 2.75 in. long,				
0.71 in. wide, and				
0.042 in. thick				
miscellaneous shapes:				
wafers 1 cm dia. × 1 mm thick	540		0.08	1.2
wafers 0.5 cm dia. × 1 mm thick	580		0.15	2.3
pellets 0.06 in. dia. × 0.06 in. thick	590		0.23	3.6
pellets 1 mm dia. × 1 mm thick	590		0.17	2.7
slabs 6 in. and 3 in. long stainless	560		0.46	7.2
Cobalt density: 8.80 ± 0.05 g/cm³			1.09	17.0
		Grand total	8.82	137.5

*Activity and power as of June 30, 1967.
†Wafers have central hole 0.070 in. in diameter.
‡One hundred and ten wafers have experimental compositions.
§Sixty-eight wafers have central hole of 0.070 in. diameter.

Approximate production costs and irradiation times for producing large quantities of ^{60}Co are shown in Fig. 7. Minimum costs are from about 10 cents/curie at 100 curies/g to about 20 cents/curie at 400 curies/g and do not change much with allowable irradiation time if at least 1 to 1½ years are allowed. These costs include estimates of all operational expenditures, including reactor operation and maintenance, components, and fuel burnup, but do not include encapsulation costs (expected to add about 2 cents/curie) nor factors related to capital, such as amortization, depreciation, and miscellaneous overhead.

The Fig. 7 curves are based on one reactor producing ^{60}Co as its major product. Quantities of ^{60}Co less than about 10 Mc/year would be considered incidental production and would be made in reactor charges designed for other products. The irradiation time would depend on the neutron flux in those charges, but the ^{60}Co cost would not be substantially higher than shown on the curves. For quantities much larger than 10 Mc/year, design of reactor charges would be influenced by the need to minimize irradiation time for the ^{60}Co product.

Capsule Material Selection

The SRL ^{60}Co program has as one of its objectives the development of satisfactory capsules of radio-

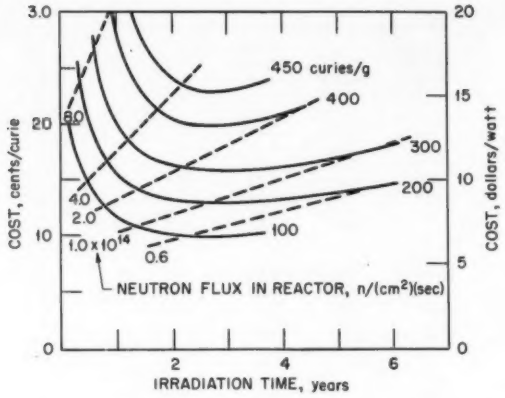


Fig. 7 Production cost estimates of ^{60}Co. (For large amounts.)

active cobalt metal contained in oxidation-resistant materials such as nickel- or cobalt-based "superalloys." Such capsules are expected to operate above 800°C and as high as 1000°C for lifetimes of 5 years or more.

Capsules must also be designed to withstand emergency conditions such as impact, thermal shock, loss at sea, fire, and external pressure. The SRL program has concentrated on normal long-term operating conditions, although limited analysis and experimentation on emergency conditions have been included.

The capsule material must be compatible with cobalt and resistant to creep, stress rupture, and in some cases, oxidation. After screening a large number of candidate materials, the alloys currently scheduled for more extensive testing are: TD Nickel, TD Nickel Chromium, Inconel* 600, Hastelloy† C, Hastelloy† X, Nickel 270, Haynes† Alloy 25, and Haynes†‡ Developmental Alloy 188. The compositions of these candidates are given in Table 5.

*Trademark of International Nickel Company, Inc.
†Trademark of Union Carbide Corporation.
‡Developmental and not marketable.

COMPATIBILITY

An encapsulating material is said to be compatible with the cobalt if (1) growth of diffusion layers and (2) rate of ^{60}Co diffusion are slow enough to prevent significant property changes and release of ^{60}Co over the lifetime of the source.

Diffusion layers often contain intermetallic layers and/or voids, which will result in property changes. As shown in Fig. 8, growth of diffusion layers is slowest in Inconel 600 and TD Nickel. The calculated widths of diffusion layers after 5 years were based on rates measured in 1-week tests at 800, 1000, and 1200°C.

Diffusion of ^{60}Co may be a more limiting factor in design than growth of diffusion layers when the capsule wall is exposed to a flowing coolant. Cobalt-60 could diffuse to the exterior, become a part of the oxidation layer, and produce a radiation hazard if carried by the coolant outside the biological shield. Data measured for the diffusion of ^{60}Co in Inconel 600 are shown in Fig. 9. Also shown are 100-hr diffusion data at 800 and 1000°C and the calculated curve of the concentration gradient after 1 year at 800°C. Both volume diffusion and grain-boundary diffusion of the ^{60}Co atoms occur, and extrapolation of the data indicates that grain-boundary diffusion may predominate in heat sources. Standard radiotracer techniques are being used to obtain diffusion data that can be analyzed by the methods of Suzuoka.[19]

Table 5 COMPOSITIONS OF CLADDING MATERIAL*

	Source	Nominal composition, wt.% Mo	Cr	Fe	Ni	Co	Mn	W	Other	Melting point range, °C
TD Nickel	Du Pont	-	-	-	98	-	-	-	ThO_2 2.2	1455
TD Nickel Chromium	Du Pont	-	20.0	-	78	-	-	-	ThO_2 2.0	1400–1430
Inconel 600	International Nickel	-	15.8	7.20	76.0	-	0.20	-		1370–1425
Hastelloy C	Union Carbide	16	15.5	5.0	54	2.5	-	4.0		1265–1345
Hastelloy X	Union Carbide	9	22	18	47	1.5	-	0.6		1260–1355
Nickel 270	International Nickel	-	-	-	99.98	-	-	-		1455
Haynes Alloy 25	Union Carbide	0.5	20	3	10	50	1.5	15		1330–1410
Haynes Developmental Alloy 188	Union Carbide	-	23	2	21	40	0.5	13		1300–1400

*Melting point of pure metallic cobalt is 1495°C.

STRENGTH

The internal pressure in a ^{60}Co capsule is determined primarily by the temperature increase of the gases present during encapsulation and secondarily from differential thermal expansion and gases released from the cobalt metal. Therefore the operating pressure of a cobalt capsule, and hence the stress in the cobalt wall, is very low and can be controlled during encapsulation by controlling the pressure during welding. For this reason superalloy materials can be used to encapsulate cobalt for operation at temperatures where the strength of these materials is generally considered low. A comparison of the approximate capsule wall thickness required to pre-

vent stress rupture or ½% creep in 5 years is given in Fig. 10 for some of the materials under consideration. These data are from the literature and will be verified, primarily by long-term tests of actual capsules.[20] The alloys TD Nickel and TD Nickel Chromium retain their strength to temperatures up to their melting points and would be the preferred alloys when strength is the main consideration. Haynes Alloy 25 and Haynes Developmental Alloy 188 also have excellent high-temperature strength. Inconel 600 has a satisfactory stress-rupture strength but may suffer excessive creep above about 950°C.

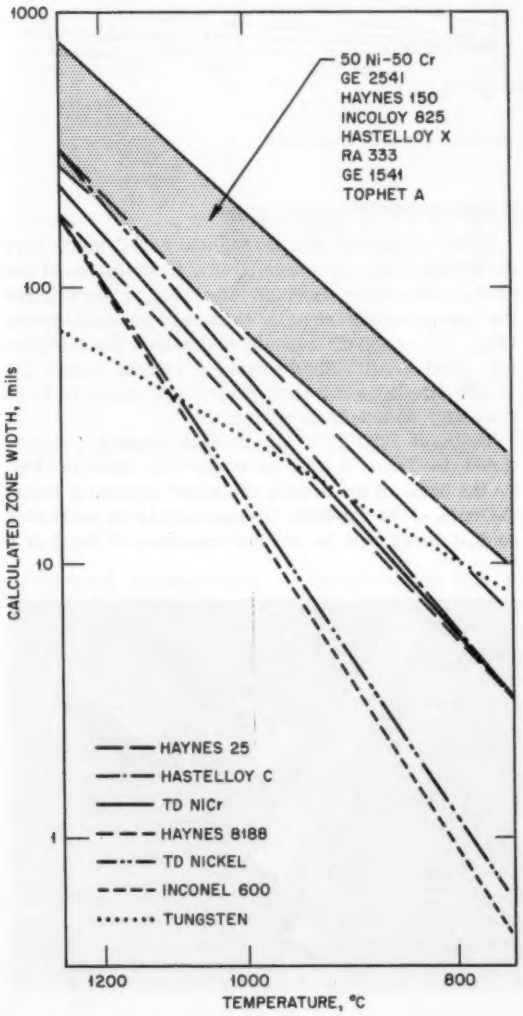


Fig. 8 Calculated diffusion-zone widths after one half-life of ^{60}Co. (Based on 168-hr anneal.)

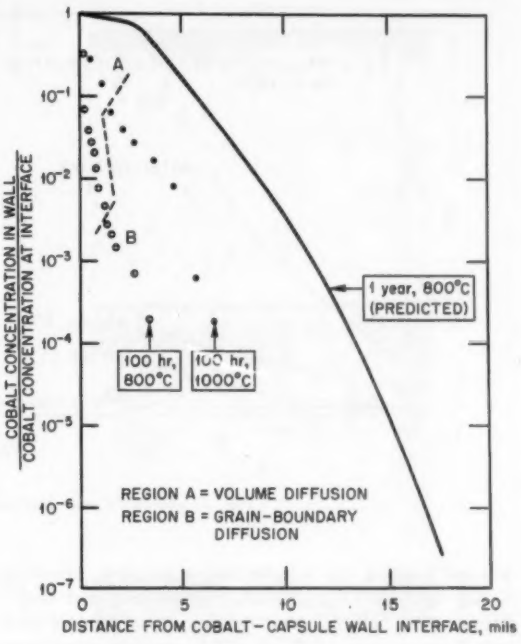


Fig. 9 Diffusion of cobalt through Inconel 600.

OXIDATION

All encapsulating alloys being considered for ^{60}Co are sufficiently oxidation resistant for normal handling in air; some are suitable for extended operation in air at heat-source temperature. Actual data from 5000-hr oxidation tests at 1000°C are given in Table 6. The alloy TD Nickel Chromium is the most resistant to high-temperature oxidation.

CHOICE OF ENCAPSULATING MATERIAL

Inconel 600 is the preferred material for experimental capsules that will operate at 850°C because of its compatibility with cobalt metal and its fabricability. One or more of the other alloys may be preferred for tests at higher temperature. Hence long-term testing of 95-mil-wall capsules of these alloys at 1000°C is planned.

Capsule Fabrication and Testing

Capsules containing irradiated and unirradiated cobalt are being heated up to 10,000 hr at typical heat-source conditions (850 to 1000°C) and examined metallographically for structural integrity, compatibility with cobalt, and oxidation resistance.

ENCAPSULATION DEVELOPMENT

Procedures for dissolving the aluminum jackets from the irradiated wafers, cleaning and gaging the

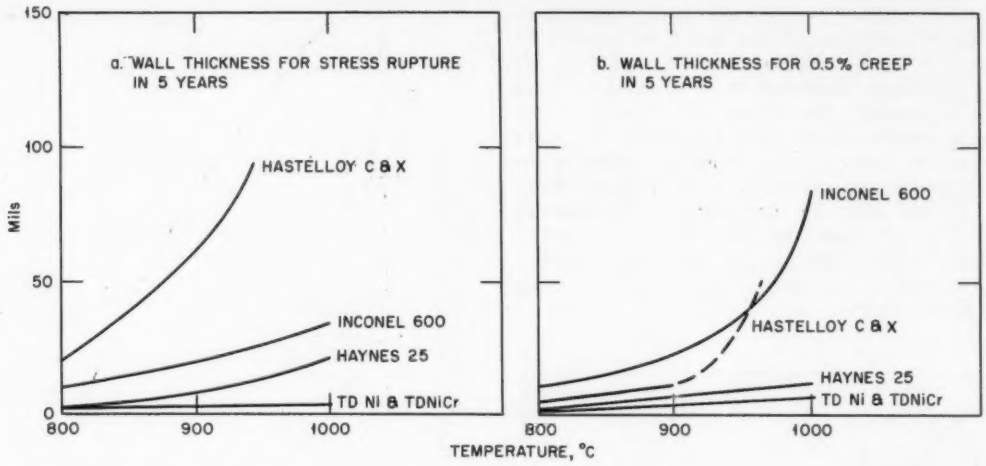


Fig. 10 Relative wall-thickness requirements for ^{60}Co capsules.

wafers, loading the wafers into a capsule, pressing a cap into the capsule after evacuating and backfilling with helium, and welding with conventional gas tungsten-arc (TIG or GTA) welding equipment have been adapted for use in the SRL High Level Caves. Whereas the wafers are loaded edge to edge in the aluminum slugs for irradiation, they are stacked face to face in the heat-source capsules (Fig. 11). In the thoria-strengthened alloys, TD Nickel and TD Nickel Chromium, the thoria dispersion segregates along stringers in the weld. This segregation may lead to nonuniform oxidation penetration in TD Nickel and a reduction of high-temperature strength in the weld of both alloys, and may ultimately require specialized closures, such as magnetic force welds. However, for purposes of the long-term "normal"-condition tests at Savannah River, TIG welding has been satisfactory.

Table 6 OXIDATION OF CANDIDATE ENCAPSULATING ALLOYS
(Still Air at 1000°C for 5000 hr)

	Thickness affected, mils		
	Surface scale	Intergranular oxidation	Zone affected by oxidation*
TD Nickel	24.4	None	Complete†
TD Nickel Chromium	2.0	None	1.6
Inconel 600	1.6	7.1	10.0
Hastelloy C	2.8	2.8	6.0
Hastelloy X	1.2	3.4	10.0
Nickel 270	30.4	Complete†	Complete†

*Zone in which either new phases were formed or preexisting phases disappeared.
†Extended throughout the 60-mil thickness of the sample (both surfaces exposed).

CAPSULE TESTING

Tests of Inconel 600 capsules at 850°C in air have been under way since April 1967. Examination of one of the radioactive capsules after 1000 hr showed that the performance of this capsule was satisfactory (Fig. 12). Another capsule maintained its integrity for 5000 hr. If other capsules remain sound for 10,000 hr, one or two capsules will continue to be tested for an indefinite period.

Tests at 1000°C in air on each capsule material listed in Table 5 will be under way early in 1968. On the basis of the results of further screening tests, the number of capsules is expected to be increased so that there will be several capsules of the 2 or 3

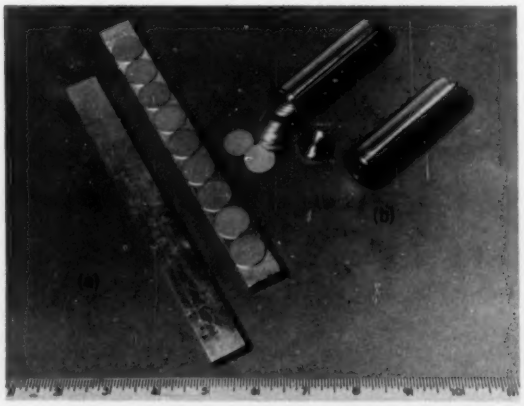


Fig. 11 Cobalt-60 wafers (a) loaded edge to edge in aluminum jackets (D bars) for irradiation and (b) stacked face to face in the heat-source capsules.

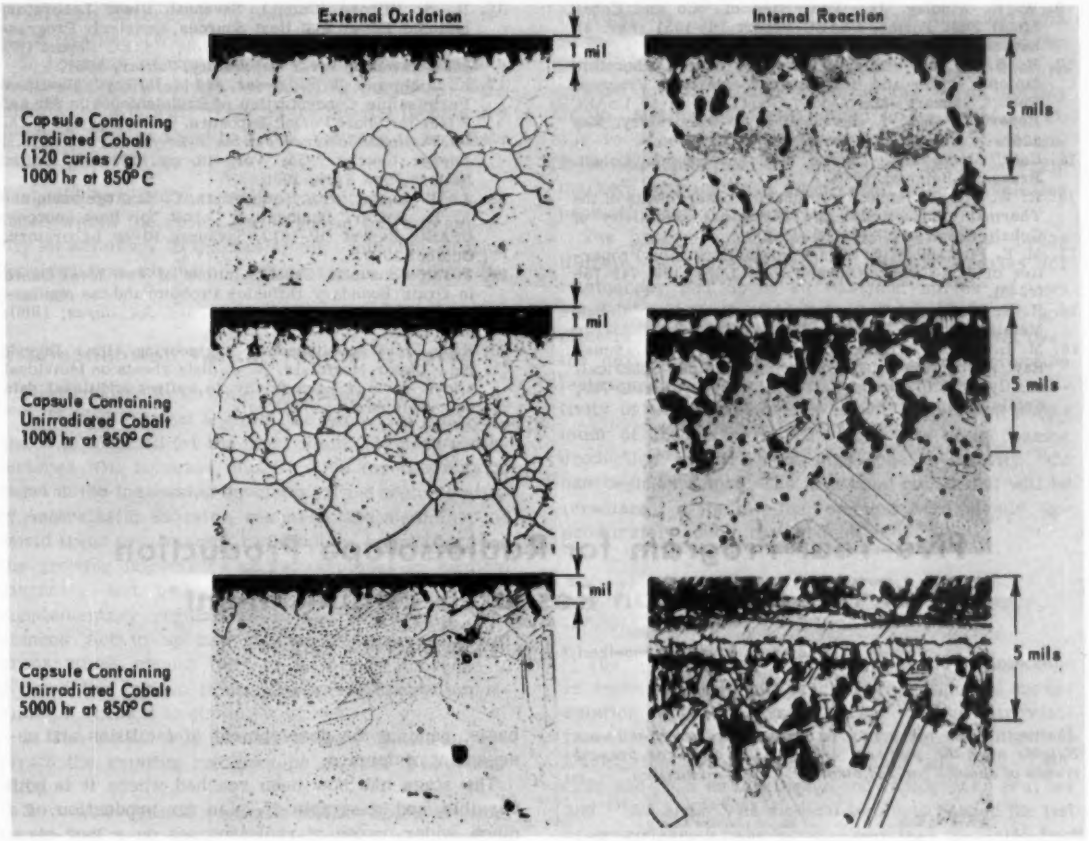

External Oxidation
Internal Reaction
Capsule Containing Irradiated Cobalt (120 curies/g) 1000 hr at 850° C
Capsule Containing Unirradiated Cobalt 1000 hr at 850° C
Capsule Containing Unirradiated Cobalt 5000 hr at 850° C
1 mil
5 mils

Fig. 12 Oxidation and compatibility in Inconel 600 capsules.

preferred encapsulating materials under test for extended periods.

Special tests of capsules to determine creep resistance, the effect of greater than normal temperatures, and the effect of defects will be conducted. For each long-term test of a radioactive capsule, a duplicate capsule containing unirradiated cobalt will be tested. The possibility of increasing the temperature limits for long-term operation by utilizing refractory metals in the capsules will also be studied in 1968. (RHL and JEC)

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Five-Year Program for Radioisotope Production at the Australian Research Establishment*

By R. E. Boyd,† U. Engelbert,† J. N. Gregory,† and G. L. Miles†

Abstract: Plans for radioisotope production at Lucas Heights over the period 1966–1971, based on present trends of demand for radioisotopes, are summarized.

Production of radioisotopes at the Australian Research Establishment began in 1960.‡ Originally emphasis was on production of radioisotopes that required a minimum of chemical or other processing, but, with the gradual improvement of facilities, the range of products has been increased. Completion of the extensions to the Isotope Production Building will enable a substantial increase in production of processed radioisotopes. Long-term planning of radioisotope production has, during the initial period, been restricted largely to the field of radiography and teletherapy sources. Production of other radioisotope sources is necessarily on a more ad hoc basis, pending the development of facilities and assessment of demand.

The stage has now been reached where it is both possible and desirable to plan for production of a much wider range of radioisotopes on a long-term basis. This article considers the program for a 5-year period ending in December 1971 and takes into account the experience of the first 5 years of radioisotope production at Lucas Heights. It also takes into account the stated policy of the Australian Atomic Energy Commission (AAEC), who approved production of radioisotopes at Lucas Heights in the following cases, where special circumstances exist or where they can be produced economically:

1. Where the radioisotope has a short half-life and it must be produced locally to satisfy domestic requirements, e.g., ^{24}Na and ^{198}Au.

2. Where importation is uneconomic because of shielding, e.g., ^{192}Ir and ^{60}Co.

3. Where production in Australia is necessary in order to ensure continuity and convenience—primarily for medical purposes; these radioisotopes would generally have short lives, e.g., ^{131}I and ^{32}P.

4. Where reactor space and surplus reactivity are available in the reactor and the cost of irradiations would be trivial in relation to the value of the radioisotope produced, e.g., ^{60}Co and ^{192}Ir.

5. For activation analysis, i.e., production of radioisotopes in situ for particular experiments, e.g.,

*Reprinted, with minor editorial changes, from Australian Report AAEC/TM 360. The authors of this report kindly brought the text up to date to reflect changes since the report was written. Unfortunately the authors' letter was received too late to be incorporated. The text as it now stands therefore must be considered as describing the program at the time of writing of the original report (November 1966).

†Australian Atomic Energy Commission, Research Establishment, Lucas Heights.

‡See also Radioisotopes in Australia, *Isotop. Radiat. Technol.*, 3(3): 270–273, 277–279 (Spring 1966).

irradiation of copper in butter, manganese in ore, and activation of superphosphate.

6. Labeling of compounds, i.e., preparation of a particular radioisotope for labeling purposes, e.g., labeling of silt with ^{198}Au, where special factors are involved such as short half-life, the need to irradiate a local material, or cost.

It should be noted that, where radioisotopes are produced within the policy stated above, there is generally an advantage to increasing the scale of production provided that adequate markets can be found for the product.

Assumptions on Which Program Is Based

The proposed program assumes that:

1. During the next 5 years, the demand in Australia and New Zealand for the most commonly used radioisotopes will increase. Justification for this may be found in the increasing development and sophistication of Australasian industry, research, and medicine; the world trend to advancing radioisotope technology; and the growing importance of radioisotopes in medical diagnosis and biological research. (The issue of supplementary regulations to the Therapeutic Substances Act to specify radioactive pharmaceutical preparations should lead to a marked increase in demand for certain items that were hitherto impossible or difficult to obtain.)

2. The AAEC will be able to some extent to penetrate the existing radioisotope market in Southeast Asia.

3. A well-defined research and development effort by the Radioisotopes Production Section is focused on improving routine products, developing new processes, and, in general, maintaining a level of technical competence in step with that of other radioisotope producers.

4. That the present High Flux Australian Reactor (HIFAR) irradiation facilities allocated to the Isotope Division will be increased by at least two hollow-fuel-element positions for fission-spectrum neutron activation and possibly two 4-volt facilities (for bulk ^{60}Co production).

Future Activities

LARGE SEALED ^{60}Co SOURCES

The sale of large sealed ^{60}Co sources is expected to continue to provide a large proportion of the revenue from radioisotope trading. Emphasis will continue to be on the production of high-specific-activity material and of teletherapy sources. It is anticipated, however, that a significant proportion of the ^{60}Co produced could be sold in the form of unsealed bulk sources.

Market assessments have shown that, although the demand for teletherapy sources was fairly constant in Australia, New Zealand, Southeast Asia, and India in the past 3 years, a steady increase in demand can be expected up to 1971. There has recently been an upward trend in other parts of the world, notably Japan, and the predicted slump in demand for ^{60}Co due to accelerator developments did not eventuate. It is expected that the Australian share in world markets will increase as a result of an active sales promotion now being carried out.

The present annual production of ^{60}Co at Lucas Heights is approximately 30 thousand curies for teletherapy and about 10 thousand curies of low-specific-activity material for gamma-radiation applications. An increase in this production level over the next 5 years is proposed. Improved activation techniques will be used to increase the specific activity of teletherapy cobalt above the present maximum of 250 curies/g. The feasibility of increasing production in HIFAR of high-specific-activity ^{60}Co has been examined. The additional cobalt that will be irradiated in hollow-fuel-element rigs should approximately double the production of ^{60}Co.

PRODUCTION OF RADIOISOTOPES OF SHORT OR MEDIUM HALF-LIFE WHICH REQUIRE LITTLE POST-IRRADIATION TREATMENT

The work involved with this type of radioisotope is concerned only with target preparation and encapsulation (where necessary), irradiation, postirradiation measurement, and preparation for dispatch. During the fiscal years 1964–1965 and 1965–1966, ^{192}Ir and ^{60}Co as level-gage and radiography sources and ^{198}Au and ^{90}Y as medical implants headed the list of unprocessed radioisotopes required (Table 1), and the demand for these products is increasing. Approximately 60 other unprocessed radioisotopes are offered in the radioisotope catalog, but the demand for these is small. The financial return in relation to the effort and resources required for production is very good in the case of ^{192}Ir and ^{60}Co, and the demand for these and the other unprocessed radioisotopes, ^{170}Tm and ^{153}Gd, is expected to rise steadily during the next 5 years. A conservative estimate would predict an increase in demand by a factor of 2 to 3, but very great flexibility in production capacity is now available.

RADIOCHEMICAL AND RADIOPHARMACEUTICAL PREPARATIONS

Preparation of radiochemical and radiopharmaceutical products involves postirradiation chemical processing of various degrees of complexity. It is usually necessary to supply the radioisotope in solution in the particular chemical form required by the user. These radioisotopes are used extensively in scientific research, industrial and field tracer experiments, and medical diagnosis and therapy. Medicine frequently demands sterile, isotonic, pyrogen-free

Table 1 SHIPMENTS OF RADIOISOTOPES IN SIGNIFICANT DEMAND PRODUCED BY THE RADIOISOTOPES PRODUCTION SECTION FROM JULY 1964 TO JUNE 1966 (EXCLUDING ^{60}Co TELETHERAPY AND LARGE GAMMA-RADIATION SOURCES)

Radioisotope	Unprocessed				Processed			
	Activity, mc		No. of units consigned		Activity, mc		No. of units consigned	
	1964–1965	1965–1966	1964–1965	1965–1966	1964–1965	1965–1966	1964–1965	1965–1966
^{192}Ir	748,600	1,161,940	59	103	-	-	-	-
^{60}Co	12,300	21,283	40	483	-	-	-	-
^{170}Tm	-	2,131	-	5	-	-	-	-
^{198}Au	3,100	2,270	47*	603†	36,100	8,505	8	17
^{90}Y	700	1,159	42*	267†	-	-	-	-
^{51}Cr	0.5	-	1	-	306,200‡	102,000‡	19	65
^{32}P	-	-	-	-	-	2,436	-	197
^{24}Na	2,700	-	14	-	800	1,687	8	45
^{42}K	40	-	2	-	120	825	6	10
^{82}Br	-	-	-	-	-	41	-	51
^{153}Gd	-	22	-	2	-	-	-	-
^{86}Rb	-	-	-	-	20	108	4	10
^{45}Ca	-	-	-	-	8	44	4	19
Service irradiations	-	-	279	139	-	-	-	-
Total	-	-	484	1,602	-	-	49	414

*Refers to shipments.
†Refers to units; unit system adopted with introduction of computer data storage, e.g., a shipment of ^{198}Au may contain up to 15 units or a shipment of ^{90}Y up to 5 or 6.
‡Substantial proportion used by AAEC in hydrological studies.

products, and, in general, the specification for purity and radioactive content is more strict for this type of product than that described in the preceding section. Some of the important isotopes for medical use are ^{18}F, ^{22}Na, ^{24}Na, ^{28}Mg, ^{32}P, ^{35}S, ^{36}Cl, ^{38}Cl, ^{42}K, ^{45}Ca, ^{47}Ca, ^{51}Cr, ^{52}Mn, ^{54}Mn, ^{55}Fe, ^{59}Fe, ^{60}Co, ^{63}Zn, ^{65}Zn, ^{82}Br, ^{85}Kr, ^{89}Sr, ^{90}Y, ^{99m}Tc, ^{131}I, ^{197}Hg, and ^{198}Au.

The details for chemically processed radioisotopes during fiscal year 1964–1965 (Table 1) refer only to materials for industrial scientific use. Medical preparations were not on the AAEC production schedule during that period. The demand for radiochemicals in the industrial and scientific fields is sporadic and highly variable in the quantities required per shipment, and for this reason there are insufficient statistics to predict the future pattern.

The previous pattern of demand for the more popular medical radioisotopes obtainable from overseas is shown in Fig. 1. These statistics were taken from the annual report of the Commonwealth X-Ray and Radium Laboratories in Australia and include the figures for New Zealand published by the National Radiation Laboratory.

Because of legal restrictions, production of radiopharmaceutical solutions did not start in Australia until July 1965. Until then, the medical profession in Australia and New Zealand was virtually without supplies of the short-lived hard gamma emitters—^{24}Na, ^{42}K, and ^{82}Br—so popular overseas. There are thus no demand statistics to give a firm indication of a future requirement in this field, but it is known definitely that they would be used if available. Long-term planning can therefore be based only on comparable consumption statistics from other countries of similar development. For radioisotopes that have been used in previous years, it can be fairly assumed that demand will increase by a factor of 2 to 3 in the next 5 years.

Some of the nearer centers in Asia are also within economic range for supply of the shorter lived radioisotopes and are expected to have some demand for AAEC products.

In general, radiochemical or radiopharmaceutical preparation requires the greatest effort in skilled manpower in radioisotope production. Therefore standard stock solutions of a given activity are produced at regular intervals in a shielded facility equipped with apparatus to deal with one or a small number of chemical processes. The economic employment of staff made possible in this method of production far outweighs the incidental waste of radioactive material. All major radioisotope suppliers use this technique for standard products.

Table 2 shows a production scheme for radiochemicals, based on predicted demands, which is possible with the design of the Radiochemical Wing, Block A, Building 23 at Lucas Heights. This scheme was designed to be flexible enough to cope with credible variations in quantity and frequency of the standard products listed and to accommodate production, at

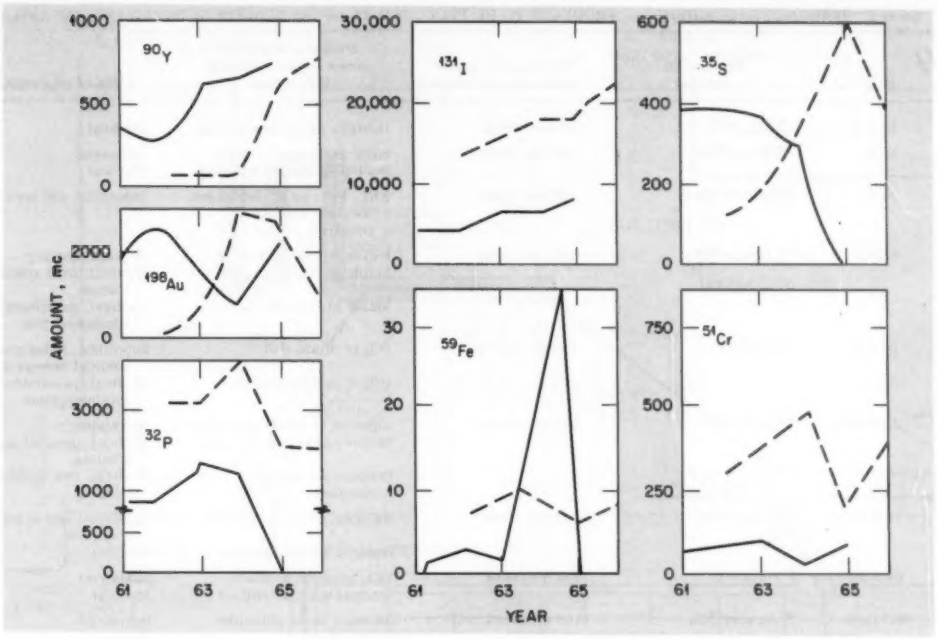

Fig. 1 Australasian usage of medical radioisotopes. Data published by Commonwealth X-Ray and Radium Laboratory (---) and Dominion X-Ray and Radium Laboratory (——).

irregular intervals, of nonstandard isotopes—^{18}F, ^{28}Mg, ^{125}I, ^{132}I, ^{99}Mo, ^{99m}Tc—as well as small amounts of fission products and very large processed sources for industrial and scientific applications.

The preparation of labeled compounds, which are becoming important in scientific research, is, with a few possible exceptions, outside the capacity of both the proposed plant and staff complement. Neohydrin labeled with ^{197}Hg (half-life 65 hr) is now in demand in Australia and is not satisfactorily available from overseas. Production at Lucas Heights should be possible after a short period of production development. Commonwealth X-Ray and Radium Laboratory estimates an initial demand of approximately six shipments per week, totaling 10 to 20 mc. The demand for compounds labeled with other very short-lived radioisotopes is small at present, but demands may arise for medical uses.

SERVICE IRRADIATIONS

The demand for radioactivation of materials supplied by clients for various purposes such as activation analysis and industrial and scientific tracer work is small. In general, this is very similar to the products described two sections above, with the client supplying the target material. Any anticipated demand in the future can be accommodated easily in present facilities.

OTHER SERVICE WORK

The principal service in this category is the importing of nonmedical radioisotopes that, for various reasons, are best obtained from overseas. In general, this includes radioisotopes which cannot be produced in HIFAR and labeled compounds of long-lived radioisotopes for which the small Australian demand does not at present justify local production (Table 3). Apart from the administrative aspects of procurement, all imported radioisotopes are inspected for safety reasons and to ensure that the import meets the customers' specification. A small part of the handling facilities available has been allocated for this service. Where sufficient demand exists it is anticipated that some of the radioisotopes will be imported in bulk and dispensed at the Research Establishment for distribution in small lots.

Other miscellaneous service work includes the loading of sources into clients' equipment, special encapsulations to order, and the disposal of replaced or spent sources.

Facilities

LARGE ^{60}Co SOURCES

The ^{60}Co handling cell and its necessary ancillaries are now in full-scale operation, and it is unlikely

Table 2 STANDARD RADIOCHEMICAL PRODUCTS TO BE PRODUCED AT LUCAS HEIGHTS BETWEEN 1966 AND 1971

Radio-isotope	Half-life	Production process	Anticipated regular batch size for stock	Probable chemical forms (other demands may arise)	Field of expected usage
^{42}K	12.5 hr	$^{41}K(n,\gamma)^{42}K$	50 mc/week	Isotonic chloride solution	Medical
^{24}Na	15 hr	$^{23}Na(n,\gamma)^{24}Na$	100 mc/week	Solid carbonate Isotonic chloride solution	Industrial Medical
^{82}Br	36 hr	$^{81}Br(n,\gamma)^{82}Br$	100 mc/week	NH_4^+, Na^+, or K^+ bromides, isotonic solution if required	Industrial and medical
^{131}I	8 days	$^{130}Te(n,\gamma)^{131}Te \xrightarrow{\beta} {}^{131}I$	1000 mc/week	Iodide in NaOH solution Iodide in $Na_2S_2O_3$ solution Iodide in isotonic buffer	Protein labeling Medical, oral administration Medical, parenteral administration
^{32}P	14 days	$^{32}S(n,p)^{32}P$	1 curie/month	PO_4 in dilute HCl PO_4 in isotonic buffer	Scientific, industrial, and medical research Medical, parenteral administration
^{51}Cr	27.8 days	$^{50}Cr(n,\gamma)^{51}Cr$	100 mc/month	Aqueous chromic chloride Isotonic chromic chloride Isotonic potassium chromate	Industrial Medical, general protein labeling Medical, red blood cell labeling
^{64}Cu	12.8 hr	$^{63}Cu(n,\gamma)^{64}Cu$	100 mc/week	Aqueous chloride solution Isotonic EDTA complex	Industrial and scientific research Medical
^{90}Y	64.5 hr	$^{89}Y(n,\gamma)^{90}Y$	100 mc/week	YCl_3 aqueous solution Yttrium silicate colloid	Industrial Medical
^{198}Au	2.7 days	$^{197}Au(n,\gamma)^{198}Au$	100 mc/week	Aqueous auric chloride Gold colloid	Industrial Medical
^{99m}Tc	6.0 hr	$^{98}Mo(n,\gamma)^{99}Mo \xrightarrow{\beta} {}^{99m}Tc$	2 curies/week	Pertechnetate generator Colloidal heptasulfide Labeled albumin	Medical
^{28}Mg	21.3 hr	$^{26}Mg(t,p)^{28}Mg$	20 mc/month	Isotonic chloride solution	Medical
^{197}Hg	67 hr	$^{196}Hg(n,\gamma)^{197}Hg$	50 mc/week	Labeled Neohydrin isotonic solution	Medical
^{47}Ca	4.7 days	$^{50}Ti(n,\alpha)^{47}Ca$	10 mc/month	Isotonic chloride solution	Medical
^{18}F	1.7 hr	$^{16}O(t,n)^{18}F$	20 mc/month	Isotonic sodium fluoride solution	Medical

that any major additions or alterations will be required over the next 5 years. The only facility demands for this aspect of the program will involve new irradiation rigs and a small increase in the number of heavily shielded transport containers.

Table 3 RADIOISOTOPES FOR WHICH PRODUCTION BY AAEC IS AT PRESENT NOT POSSIBLE OR DESIRABLE AND WHICH WILL NORMALLY BE IMPORTED

Radioisotope preparation	Reason not produced by AAEC
Accelerator-produced neutron-deficient isotopes	Australian accelerator resources very limited
Fission products	Expensive and complex plant required but may be justified in some cases, e.g., ^{99m}Tc
Tritium and other radioactive gases; tritiated water	Supply assured from overseas; no transport problems
Labeled compounds of ^{14}C, ^{3}H, ^{35}S, ^{75}Se, ^{36}Cl, ^{131}I	Extensive resources and highly trained staff required and not normally economic
Special sources (e.g., ^{3}H targets, Mössbauer)	Very small Australian demand
Processed radioisotopes not produced by AAEC	Many processed radioisotopes will be imported, especially those of long half-life

NONPROCESSED RADIOISOTOPES OF SHORT AND MEDIUM HALF-LIFE, SERVICE IRRADIATIONS, AND MISCELLANEOUS SERVICES

All facilities necessary to achieve the program outlined above are either in existence or in an advanced state of design or construction. Figure 2, a diagram of the materials flow when all these facilities are in operation, demonstrates that shielded cells 1–14, on the northern part of the radioisotope handling bay in Building 23, and now used exclusively for manufacture of unprocessed radioisotopes, will also be required for the initial postirradiation handling of activated substances, which will be chemically processed in Building 23A. This requirement must be accounted for in any assessment of the maximum activity allowable in the cells. In order to avoid undue handling problems, some increase in the present shielding of these facilities must be incorporated in the building program for the period under review.

These facilities will cope with practically all the demands made by service irradiations and the other

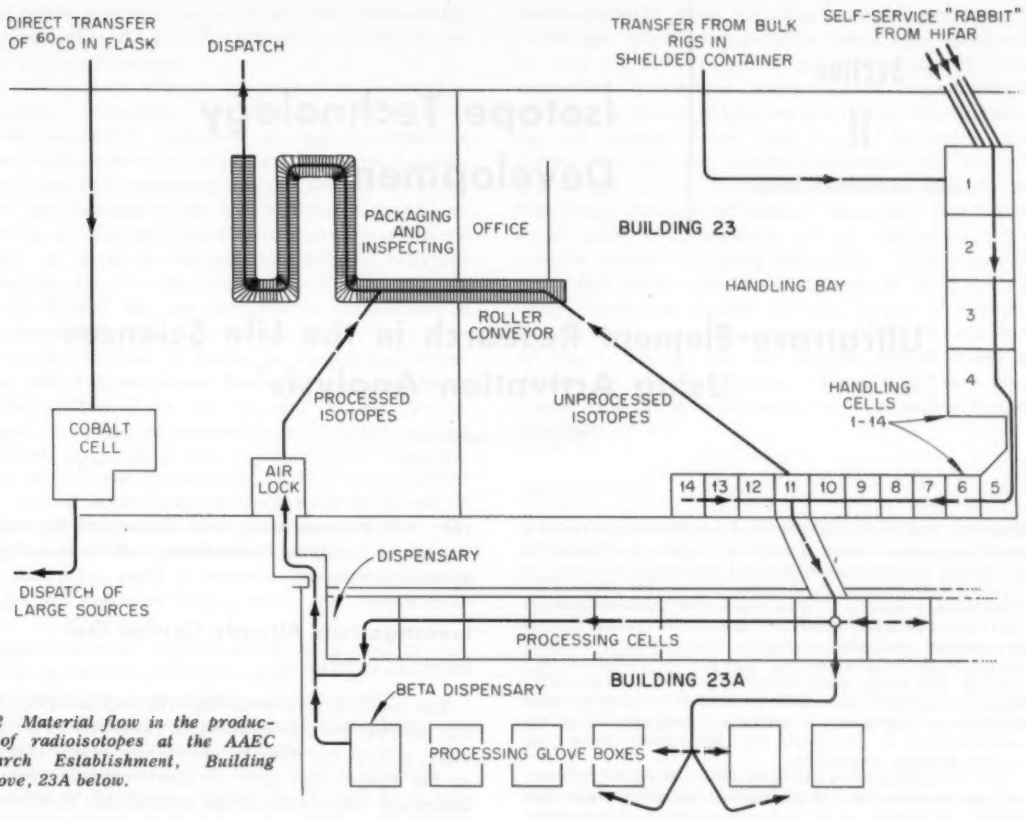

Fig. 2 Material flow in the production of radioisotopes at the AAEC Research Establishment, Building 23 above, 23A below.

miscellaneous service activities, including dispensing of gaseous radioisotopes which have been imported in bulk.

RADIOCHEMICAL AND RADIOPHARMACEUTICAL PREPARATIONS

The radiochemical facilities in Building 23A will be capable of handling the program envisaged for the next 5 years.

Research and Development

A definite proportion of staff effort, materials, and facilities of the Radioisotopes Production Section is set aside to improve the quality of present products and processes and develop new products for which demands arise and which can be made using local resources.

A general program of research and development has been drafted and is now operational. In the development of new products, the program favors, in principle, the short-lived radioisotopes. It is divided into the following categories:

Chemistry. Projects in this field are directed toward development of techniques to produce radiochemical compounds and for routine control of certain quality aspects. Rapid and reliable chemical separation methods, with the object of producing carrier-free radiochemicals that are the product of other than (n,γ) activation reactions or daughter products, are a highlight in this research.

Physics. The physics research program is devised to develop suitable activity measurement techniques for the determination of the radioactive strength and radioisotopic purity of products. It also is concerned with a fuller understanding of activation processes and mechanisms in reactors or other activity inducing devices.

Engineering and Metallurgy. Effort in this field is directed toward development of more efficient activation techniques in HIFAR, improvement of transport and handling-container designs for radioactive materials, safe and efficient encapsulation methods for radiation sources (in particular those with low penetrating power), and shaping of metallic radiation sources, notably ^{60}Co. (MG)

Section

II

Isotopes and Radiation Technology

Isotope Technology Development

Ultratrace-Element Research in the Life Sciences Using Activation Analysis*

By G. W. Leddicotte†

Abstract: The method of activation analysis stands out as a significant achievement in nuclear technology. The ability to identify and determine most of the stable elements by forming their radioactive isotopes in a nuclear particle irradiation has made activation analysis a foremost analytical tool. For many elements, it is more sensitive than conventional analytical methods. The unique decay processes of the induced radionuclides and the radiations they emit as they decay make it very specific. Contamination during an analysis, frequently a problem in other analysis methods, is negligible in activation analysis unless the contamination is introduced into the sample before the nuclear particle irradiation.

The discussion presented here shows, in a general manner, the present state of the method for using trace elements in problems in life-sciences research. The applications described reflect some areas of immediate interest but are by no means exhaustive.

Biological and medical research advances during the last few years have brought about a new appreciation of the action of the chemical elements in living systems. At least 60 elements in low concentrations, i.e., in quantities equal to or less than 1 part in 1000,‡ have been detected in bacteria, fungi, higher plants, animals, and man. It is therefore no longer sensible to treat elements in these concentrations as being "just" contaminants in matter. Instead, their presence must be viewed as parts of a single intricate system or of an infinite number of chemical systems completely organized or interrelated. An intelligent understanding of the functions of these systems could very well suggest many new avenues of approach to biological problems in medicine, nutrition, and environmental science.

Investigations Already Carried Out

MEDICAL RESEARCH

The need for understanding trace-element functions has already stimulated medical researchers to speculate and to hypothesize concerning trace elements in the cause and cure of many chronic diseases. Hence any exploration of the metabolism of the chemical elements should help explain their functions in the biochemistry, physiology, and pathology of man. Already, some researchers[1] have found that all measurable biological activity in living cells can be lost when the element is split from the protein residue of the metallo-enzyme contained in the cells. It has been shown that traces of iron, copper, and molybdenum, as integral parts of the flavoprotein enzymes, have established functions in the oxidation–reduction physiology of cellular metabolism[2,3] and that, in the synthesis of ascorbic acid in the liver, cobalt or manganese act only as catalytic cofactors and are probably unessential parts of the enzyme molecule.[4] Traces of copper have also been related to ceruloplasmin activity,[5] and manganese above the trace level appears to be essential for cytoplasmic metabolism in mitochondria cells.[6]

Great interest already exists in understanding the biological aspects of metal-binding agents and their relation to human hypertension. As early as 1960, Schroeder,[7] Seven,[8] and Johnson and Seven[9] reported data relating antihypertensive agents to metal-binding agents and showed that urine from hypertensive patients contained increased amounts of many trace elements. The last two papers also suggested that additional knowledge of metal chelates in the body

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‡Although the emphasis here suggests that low elemental concentrations are equal to or less than 1 part in 1000, it is more correct to categorize elemental concentration as: major, 1% or more; minor, 0.1 to 1%; micro, 0.01 to 0.1%; trace, 0.001 to 0.01%; and ultratrace, <0.001%.

might prove that deficiencies of certain trace metals or the retention of others by specific body organs can cause chronic diseases.

To relate trace-element concentrations to the biochemistry, physiology, and pathology of a living system, normative distributions must be available to the health-sciences-oriented researchers. In 1960, Tipton[10] reported extensive work on the determination of the concentrations and distributions of trace elements in normal and diseased human tissues. Koch et al.,[2] in work on cellular metabolism, also reported that Ga, Fe, Mg, Al, Cd, Cr, Co, Cu, Mo, Ni, Sn, Ti, V, and Zn are involved in normal human physiological functions. In a later study[11] they gave data on the Cu and Zn levels in the plasmas of patients with lymphomatous diseases and reported on the distribution of Cd, Cr, Cu, Fe, Pb, Mg, Mo, Ni, Sn, and Zn in tissues obtained from biopsies or from autopsies of patients with lymphomatous diseases. Olson et al.[12] and Addink and Frank[13] first reported data on zinc metabolism in tissues and blood and its relation to cancerous and noncancerous states. Although such early investigations as these pointed out that anomalies exist in elemental concentrations in normal and diseased tissue, more work needs to be done.

Typical of other studies being carried out or considered by medical scientists are:

1. Role of trace elements in arteriogenesis
2. Role of copper and manganese in hepatalenticular degeneration (Wilson's Disease)
3. Magnesium deficiency in chronic alcoholism
4. Role of Ni, V, Al, Rb, Cd, B, Ce, and Cs in biological functions, with interest in their relation to (*a*) protein complexes of hormones and vitamins, (*b*) activation and inactivation of enzymes, (*c*) exchange of O_2 and CO_2, and (*d*) other physiological functions
5. Role of any element (other than Na, K, Mg, and Ca) in hypertension, ulcers, and gallbladder disease
6. Individuality of human serum (for identification)
7. Assay and correlation of enzyme patterns of aerospace candidates (under simulated conditions) for metabolic criterions in space
8. Relation of acceptable foods and their metabolism to space health requirements and/or personal hygiene during long-term space habitation

In addition, other investigators are beginning to consider the clinical significance of trace Sb, As, Ba, B, Br, Cr, Co, F, Ga, Pb, Li, Hg, the rare earths, Sc, Ag, Sr, Sn, Ti, Th, W, and Zr.

NUTRITION RESEARCH

Laboratory studies have shown that trace elements affect animal nutrition, and the number of trace elements considered essential increases as more studies are carried out. Various specific beneficial nutritional effects have been shown to stem from trace elements in the diet. For example, Schroeder[14] found that growth and survival rates were impaired in rats and mice raised in an environment strictly controlled to prevent trace-element contamination and with a known chromium deficiency. Supplementing the animals' diets with 5 μg of chromium improved growth and survival. Schwarz and Merz[14] found that trace chromium also aided insulin in improving glucose utilization. Baumann[14] found that small amounts of selenium (<5 μg) combatted white muscle disease in sheep and cattle. Frost[14] speculated that small amounts of arsenic in the diet may offer protection against certain forms of cancer. Other researchers have concluded that small amounts of Cu, I, Co, F, Mn, and Zn play specific roles in animal or human nutrition. Many other trace elements may prove to be beneficial in animal or human nutrition.

AGRICULTURE

An understanding of the function of trace elements is important also in agricultural research. Most studies of plant nutrition and metabolism pertain to understanding what plants need most for their best growth and how they assimilate the needed materials. Small concentrations of the elements Cu, Mn, and Zn have been found to be essential to plant growth. Other studies have shown that even though K, Rb, and Cs are chemically similar, plants will metabolize only K.

Although plants are bred to resist the pathogen of the moment, Nature is constantly changing the microbial population by mutation. As a result, an understanding of the relations between the different elements and the chemical changes they undergo within the plant cells is essential to the determination of whether the plant can be made to resist disease. A general breakthrough in the control of plant disease is yet to come, and trace elements as chemical controls for plant diseases need to be investigated.

Likewise, in order to predict what new classes of chemicals might be valuable as herbicides or as preventatives of field-crop diseases, the deleterious effects these chemicals have on human physiology must be investigated. Many of the elements used in such chemical agents, even though present in concentrations as small as 1 ppm or less, are considered to be toxic to both animals and humans.

Diseases in plants appear to have some similarity to chronic diseases in humans, and an understanding of anomalies in trace-element distributions and their role in these plant diseases could well be the key to understanding similar conditions in humans. In many other areas in agricultural science, an understanding of the trace-elements' function is required to obtain an improved product. The efficiency of rations for proper animal nutrition, the quality of animal fur and hides for clothing manufacture and shoe leather, and

the life cycles of insects are other areas that are somewhat dependent on a knowledge of trace elements.

ENVIRONMENTAL SCIENCE

The federal government continues to emphasize major research efforts in understanding the biological effects of pesticides and water quality and in regulating the disposal of waste to air and water environments. All these activities require an extended knowledge of the elemental compositions of the components involved.

Techniques for Determining Low Concentrations of Elements

At least 15 methods of analysis exist for the determination of elements, whether they appear in a sample system in macro concentrations or in ultratrace amounts. The methods vary in their specificity, sensitivity, and precision. The examples that follow make possible a comparison of some of these methods for determining trace elements in the health sciences.

Emission spectroscopy, flame spectrophotometry, colorimetry, and amperometric titrations were used to obtain data on the concentrations of the elements investigated in the examples cited above. In general, any of these methods is capable of determining minor or micro concentrations of almost all the elements; however, many of them lack sensitivity for a precise and accurate determination of many elements present in trace and ultratrace concentrations. For example, Tipton[10] used emission spectroscopy to determine 24 elements in at least 29 different human tissues. However, only 10 of the elements which had concentrations greater than 10 ppm could be determined with reasonable precision; the other 14 were detected in some tissues, and their concentrations were usually reported as "less than" values (e.g., <1.1 ppm Mo; <0.13 ppm Co; <3 ppm As; and <1.5 ppm V). In another investigation[15] X-ray spectrochemical analysis was used to analyze human blood serum for many elements. Of the elements sought, 29 elements—including Mn, Co, Ni, Cd, and I—were not observed at all, and 19 others were observed in only a few samples; only 18 of the elements were detected in all samples.

Analytical results such as these indicate that the sensitivity of the method used did not approach the concentration levels of many of these elements in the tissues analyzed. Hence any method for obtaining information on elemental concentrations for use in a life-science investigation must have the capability for making a significant determination of elements in the ppm to ppb range. The method also should be able to give accurate and precise data about the element when its concentration is greatly in excess of this quantity. Simultaneous analysis of as many elements as possible in the same sample specimen is also desirable. Further, the method should allow the processing of many samples rapidly with a minimum of physical manipulation between sample preparation and the final tabulation of analytical results.

The methods of gas chromatography, polarography, emission spectroscopy, atomic absorption, mass spectrometry, and activation analysis all have the ability to determine elements in concentration ranges below 0.1%. However, of these, activation analysis has the greatest advantage in that essentially all the elements can be so determined with an accuracy of better than ±5%, and many elements can be determined simultaneously.

Determination of Minor and Ultratrace Elements by Neutron Activation Analysis

Since good reviews of the principles of neutron activation analysis have appeared in the literature,[16,17] further description of the technique is unnecessary. Each of these reviews emphasizes the sensitivity and specificity of the method as well as some of its advantages and disadvantages.

Neutron activation analysis has been used to determine about 70 of the elements from milligram to submicrogram amounts in many different sample materials. Some use of the technique has already been made in biological and medical research.[18–21] However, these investigations have been concerned only with the determination of selected elements, and the potential of activation analysis techniques in understanding the role of trace elements in these research problems has not yet been fully realized.

Many activation analyses can be completed, without the use of radiochemistry, by gamma-ray scintillation spectrometry—a highly developed radiation-detection system that permits an analyst to obtain a substantial amount of data in one measurement of the radioactivity of an activated specimen. The digital pulse-height spectral data obtained from the gamma-ray scintillation spectrometer are in a form readily adapted to processing by a computer. Computer-compatible programs for analyzing the data so obtained from activation analysis are already being used. The computer-integrated approach is rapid and provides an accurate and reliable automatic data reduction system that uses a number of statistical tests to check the accuracy of the data and the resulting calculations. Analytical data and other characteristics of the sample can be stored for correlative studies.

Activation Analysis Applications in Life-Sciences Research

Because it is an excellent, specific, and sensitive method of analysis, activation analysis is receiving

growing attention from life-science researchers as the most practical analytical tool for detecting and measuring concentrations of trace elements. The examples given below show the present scope of this work and emphasize the technology that can be followed to complete the assay of such sample material.

TRACE RARE EARTHS IN BONE ASH

Data on trace rare-earth elements obtained from the assay of an irradiated bone ash sample whose components were separated by ion exchange are shown in Table 1. The ash was dissolved in a mixture of mineral acids with inactive lanthanum and cerium carrier solutions, after which the rare earths were precipitated as hydroxides by the use of NaOH. The hydroxide precipitate was dissolved in HCl and the solution transferred to a Dowex-50 ion-exchange-resin column. The elements were eluted from the column with 5% ammonium citrate buffer solution at a *p*H of 3.26, and the radioactivity of each eluted fraction was measured.

Table 1 TRACE RARE EARTHS IN BONE ASH

Element	Concentration, ppm	Element	Concentration, ppm
La	0.27	Tb	0.0014
Sm	0.009	Dy	0.001
Eu	0.20	Ho	0.50
Gd	0.00	Er	2.20
Lu	0.08	Tm	1.30
Y	0.04	Yb	1.30

TRACE ELEMENTS IN ANIMAL TISSUES

The range of data obtained in the activation analysis of many samples of human blood, bone, and soft tissues (lung, liver, pancreas, intestine, spleen, and kidney) is shown in Table 2. Each specimen was ashed, irradiated for a given time, and then processed radiochemically. Many of these data have been used by Tipton[10] in her later correlations of the distributions of trace elements in the human body.

In more recent work, protein-bound iodine in blood and urine, in concentrations ranging from 0.001 to 1.0 ppm, has been determined by neutron activation analysis. The induced ^{128}I has a half-life of 25 min. This is sufficiently long for the sample to be processed by a radiochemical separation method that employs solvent extraction and precipitation techniques. Gamma-ray spectrometry was used to complete the analysis. This work is soon to be extended to the analysis of field crops for protein-bound iodine.

Activation analysis is now being used in medical research problems concerned with the high incidence of dense bones and of thyroid disease seen in Midwesterners who have been long exposed to richly mineralized water and milk. The ability of bone and thyroid tissue to concentrate elements selectively suggests that excessive ingestion of mineral contaminants may protect or damage each tissue. Symptoms of osteoporosis are found in patients with dense bones although sporadic hypocalcemia is also found. Hence, to understand the relation between trace-element distributions and the atomic structure of disease, biopsy specimens of bone and parathyroid tissues are being examined by a nondestructive activation analysis technique. Short irradiations (4 min) at a moderate flux [about 10^{12} n/(cm^2)(sec)] and immediate assay of the irradiated specimen by gamma-ray spectrometry have shown that distribution anomalies exist in each type of sample, particularly for copper and manganese. Concentrations of each of these as small as 1 ppb and as large as a few ppm have been observed. There is some preliminary evidence that aluminum and vanadium concentrations in diseased parathyroid tissues are higher than in normal.

Table 2 TRACE ELEMENTS IN BLOOD AND SOFT TISSUES

Element	Tissue	Concentration, ppm
Cl	Blood	100
Co	Soft tissues	0.1 to 1
Sb	Soft tissues	0.01 to 1
As	Soft tissues	0.001 to 1
Br	Soft tissues	1 to 10
Cd	Soft tissues	<0.1
Cs	Soft tissues	1 to 10
Cu	Soft tissues	0.1 to 100
Na	Soft tissues	0.1 to 1
Rb	Soft tissues	1 to 10
Se	Soft tissues	0.001 to 1
Sr	Soft tissues	1 to 10
Zn	Soft tissues	1 to 100
Zr	Soft tissues	1 to 10

TRACE HEAVY METALS IN PHARMACEUTICALS

In the last year or two, an effort has been made to obtain significant information about the heavy element content of the raw materials used in the manufacture of drugs and pharmaceuticals and of the final products produced from these materials. Some of the data obtained in this application of activation analysis are summarized in Table 3. Most of the analyses of the

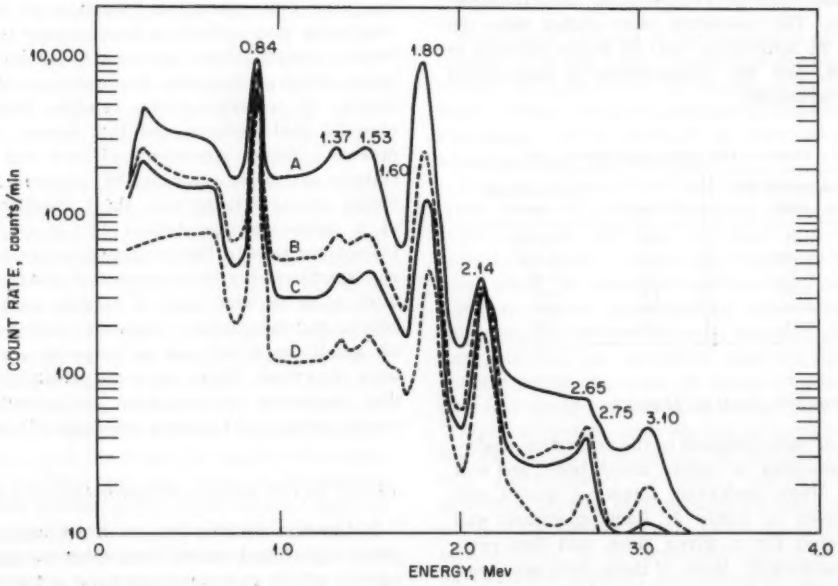
Table 3 HEAVY ELEMENTS IN DRUGS AND PHARMACEUTICALS

Material	Concentration, ppm						
	Mn	Cu	Na	As	Sb	Au	Br
Acetophenetidin	0.05	0.3	0.8				
Acetylsalicylic acid	0.008	0.1	5.0				
Benzoic acid	0.04	0.3	2.7				
Calcium Carbonate		2.1	9				2.0
Methyl salicylate	0.01	0.005	0.04				0.009
Riboflavin	0.04	0.2	3	0.05			
Adrenalin	1.9	0.5	3.8	1.4	0.7	0.03	
Vitamin C	0.07	0.06	1.1	0.05	0.1	0.01	

irradiated materials were completed by the use of radiochemistry and gamma-ray spectrometry.

MICROELEMENTS AND THE PROPERTIES OF SOILS AND PLANT MATERIALS

The use of neutron activation analysis to obtain systematic knowledge of the properties of trace elements in soils and vegetation was initiated by Bate and Leddicotte[22] in 1961. The data presented in Fig. 1 and Table 4 are typical of those obtained in a series of assays made to show that this method of analysis can be useful in determining biogeochemical requirements for ecology systems and in determining the mechanisms responsible for biological relations of soils and vegetation to human health. Figure 1 is particularly interesting because it shows the potential of the use of very short irradiation times — 10 sec in this instance. A considerable amount of information on the trace-element composition of pine needles was obtained from these analyses. When the data were placed on a quantitative basis, ppms or less of each of these elements were observed. Table 4 shows more specific information obtained in the assay of various soil types for trace cesium. This use of activation analysis has been extended to include many other studies on the microelements in soils and vegetation. For example, one study carried out recently on a selected group of cured tobacco samples showed an arsenic content, resulting from pesticide treatments, that ranged from 7.8 to 23.4 ppm. Opium, cranberries, and walnuts have also been satisfactorily



Decay curve	T_0 plus	Energy, Mev	Radionuclide	$T_{1/2}$
A	30 sec	3.13	^{37}S	5.04 min
B	6 min	3.10	^{49}Ca	8.8 min
C	10 min	2.98	^{56}Mn	2.58 hr
D	40 min	2.75	^{24}Na	15.0 hr
		2.65	^{56}Mn	2.58 hr
		2.16	^{38}Cl	37.3 min
		2.13	^{56}Mn	2.58 hr
		1.81	^{56}Mn	2.58 hr
		1.78	^{28}Al	2.27 min
		1.53	^{42}K	12.8 hr
		1.37	^{24}Na	15.0 hr
		0.845	^{56}Mn	2.58 hr
		0.834	^{27}Mg	9.45 min

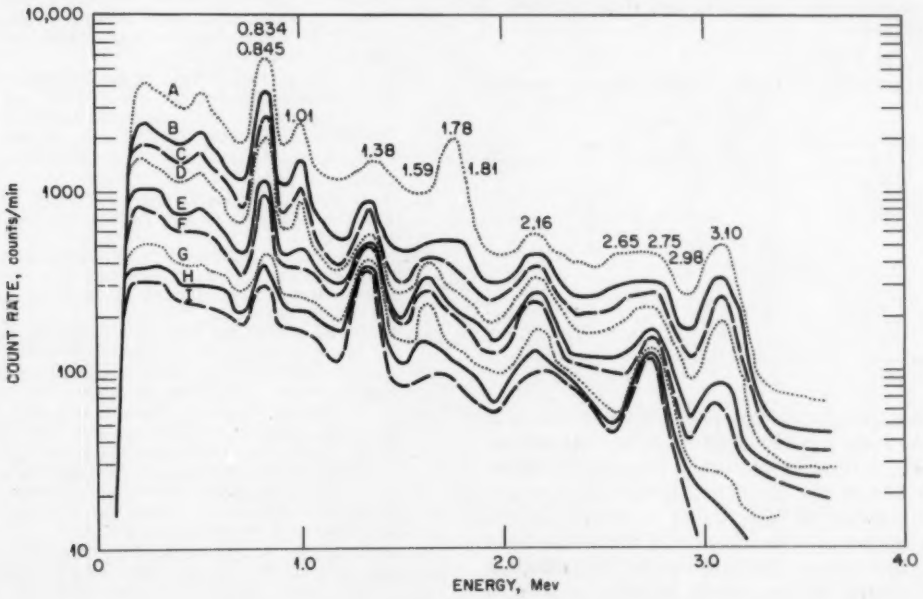
Fig. 1 Gamma-ray decay spectrum of radioactivated pine needles. Flux, 3.5×10^{13} n/(cm²) (sec); irradiation time, 10 sec.

Table 4 TRACE CESIUM IN SOILS

Soil type	Cs concentration, ppm				
	Surface	4 in. depth	7 in. depth	19 in. depth	24 in. depth
Sequoia	1.5	2.0	4.0	4.8	
Monongahela	2.8	3.4	4.2	6.3	
Landsburg	2.0	2.5	3.1	3.3	3.1
Fullerton	2.5	3.0	4.3	7.2	7.1

analyzed for trace-element contents by activation analysis methods using gamma-ray spectrometry and radiochemistry.

Studies currently under way involve (1) the study of aging in plants, (2) the relation of trace-element distributions to plant diseases, (3) the identification of plant species, and (4) the uptake of nutrients by various types of plants. Preliminary data obtained in the analysis of oat plants show that there could be a correlation between the loss or increase of trace sodium, manganese, or copper with the plant growth. In the plant-disease and plant-identification studies, early data show anomalies in trace-element distributions which appear to be useful for comparisons. The nutrient study is providing information on the uptake of various elements—Cu, Mn, V, Co, etc. This work will be extended to include studies on animals fed with these materials. It is expected that many other studies of this type will be initiated. For example, considerable interest exists in understanding the metabolism and fate of pesticides in plants and animals, and a program involving activation analysis



Decay curve	T_0 plus	Energy, Mev	Radionuclide	$T_{1/2}$
A	5.5 min	3.10	^{49}Ca	8.8 min
B	12.0 min	2.98	^{56}Mn	2.58 hr
C	17.0 min	2.75	^{24}Na	15 hr
D	22.0 min	2.65	^{56}Mn	2.58 hr
E	33.5 min	2.16	^{38}Cl	37.3 min
F	42.0 min	1.81	^{56}Mn	2.58 hr
G	65.0 min	1.78	^{28}Al	2.27 min
H	97.0 min	1.59	^{38}Cl	37.3 min
I	157.0 min	1.38	^{24}Na	15 hr
		1.01	^{27}Mg	9.45 min
		0.845	^{56}Mn	2.58 hr
		0.834	^{27}Mg	9.45 min

Fig. 2 Gamma-ray decay spectrum of neutron irradiated spring H_2O. Irradiation time, 10 min; flux, 8×10^{11} n/(cm^2) (sec); volume, 20 ml.

technology will soon be started to monitor food and people for chloride-based pesticides.

ENVIRONMENTAL SCIENCES

Emphasis is now being given to the use of activation analysis as a general analytical tool to obtain much-needed information about unusual, as well as common, elements in extremely low concentrations in water, soil, stream sediments, dusts, and similar environmental samples. Figure 2 is a typical presentation of data obtained in one such application. In this instance, a water specimen was irradiated for 10 min, and the sample activity was counted periodically on a gamma-ray scintillation spectrometer. The decay spectrums provided considerable information on the concentration of the elements identified. When this information and that obtained in subsequent analyses were quantized, data typified by that shown in Table 5 were obtained.

Table 5 ULTRATRACE ELEMENTS IN FRESHWATER SPRINGS

Element	Concentration, ppb		
	Sample 1	Sample 2	Sample 3
Cu	2.	11	5
Mn	0.4	37	9
Cl	2400.	900	800
Al	6.	620	1500
Fe	220.	240	110
Zn	17	16	0.5
Se	2	2	1
Th	0.2	0.3	0.1
Co	0.2	0.2	0.1
Cs	0.07	0.05	0.05

The significance of data presented in Table 5 is that it consists of determined values unobtainable by most water analysis laboratories. Since the activation analysis method can show the presence of many elements in addition to those listed in Table 5, it provides a means of determining if a water source is suitable for human consumption or an industrial process system. A large-scale program is soon to be undertaken at the University of Missouri to determine the trace-element distribution in Missouri water systems and to relate their distribution to the incidence of various chronic diseases. Similar programs are planned in other areas of interest in environmental science. (FJM)

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Design and Fabrication of a Low-Background-Radiation Shield*

By Harold Zallen† and Thomas R. Dixon‡

Abstract: The design and fabrication details of an easily constructed lead shield for a scintillation counter are described. The shield, which houses the scintillation detector in a horizontal position, features a ball-bearing steel-rod track suspension by which the heavy shield door can be opened or closed easily by fingertip pressure. The body of the shield is made up of cast and machined lead rings to permit easy construction as well as relocation without the use of a hoist.

The heavy weight of the lead shield for a scintillation detector presents a problem when the shield must be moved, especially to an above- or below-grade floor in a laboratory where elevators or hoists are not available. Likewise, opening or closing the heavy door or plug of some shields becomes a problem, especially for women laboratory employees. The shield described here is used in a horizontal position. It features a multiple lead-ring construction and a door that operates on a ball-bearing steel-rod-track suspension, which permits opening and closing of the heavy lead door with fingertip pressure.

The basic lead-ring design was first used by the Nuclear Measurement Corporation in a vertical-position shield with a pulley arrangement and suspended plug (or door). The shield described here is made up of 15 lead rings and two solid lead disks at the closed end (Fig. 1). Each ring has a 15.25-in. outside diameter, a 6.125-in. inside diameter, and a 1.5-in. thickness. The two solid disks are 15.25 in. in diameter and 1.5 in. thick. The same basic mold is used to cast all the rings, and their faces and those of the disks are milled flat to ensure a close fit. The rings and the disk are drilled to accommodate four threaded rods that hold the assembly together. A wedge cut from one of the rings has a groove machined into one of the milled surfaces to provide a passageway for the scintillation detector coaxial cable. A small handle attached to this wedge facilitates removal from the shield, and a light coat of silicone grease prevents the wedge from sticking. The length of the shield can be conveniently altered by adding or removing the desired number of lead rings.

The door of the shield is a lead plug 6 in. in diameter and 4.5 in. long. It has a cantilever suspension and rides on ball bushings on steel-rod tracks (Fig. 2). These ball bushings and hardened steel-rod tracks are commercially available (Thomson Industries Inc., Manhasset, N. Y.). Four bushings, two for each side, are press fitted into the base of the carriage. These

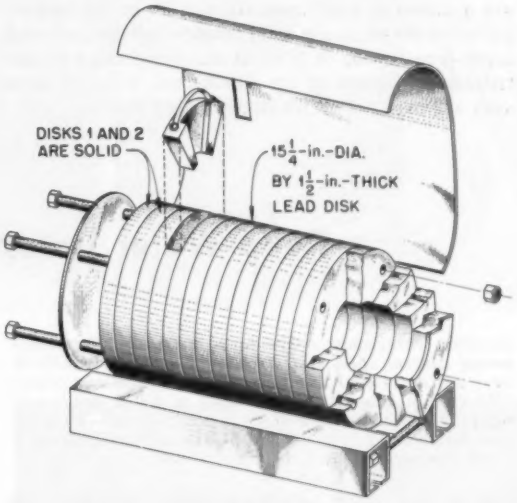

Fig. 1 Ring-type low-background-radiation shield.

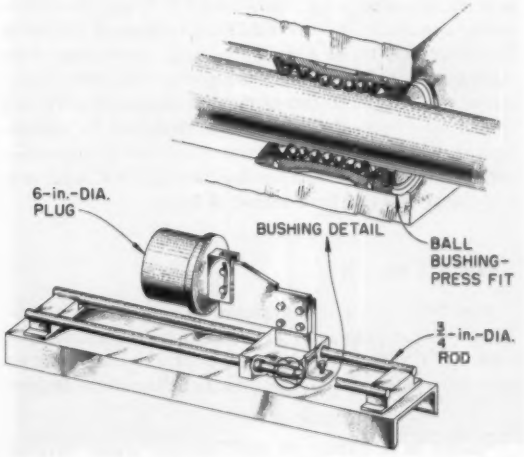

Fig. 2 Sliding door and track assembly.

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bushings, made especially for sliding linear motion, have oblong circuits of balls that permit the load to be rolled freely along the shaft on the balls. Thus the heavy door moves easily with very little effort.

Virgin lead was used to make the shield and the door, data for which are

Total volume of lead	3820 cu in. (62,000 cm^3)
Total weight of lead	~1600 lb (710 kg)
Wall thickness	4.5 in. (~11.5 cm)

The outer wall and the closed end of the shield are covered with $^3/_{16}$-in.-thick aluminum sheet, and a tubular liner of appropriate materials, e.g., Cu–Cd, can be inserted inside. The shield and the door suspension are mounted on steel channels, and the entire assembly is mounted on a heavily constructed channel- and angle-iron bench, 68.5 by 28 in., which helps to distribute the weight of the shield over a larger floor area and thus reduce the floor-loading factor.

A 2- by 2-in. NaI(Tl) scintillation crystal has been used with the shield to count environmental samples (rain, earth, etc.). A Marinelli-type beaker made of thin sheet metal, coated with a protective material, and fitted with a tight snap-on lid contains the sample and fits snugly around the crystal during counting. Perforated cylindrical paper food containers have also been used to hold small laboratory animals, i.e., rats and mice. (There is room between the door and the end of the crystal for these containers.)

The Technical Measurements Corporation (TMC) Gammascope 101 and the Nuclear Measurement Corporation Automatic Counting System (ACS-10) have both been used as analyzers. A TMC preamplifier was used with the 2- by 2-in. NaI(Tl) crystal and photomultiplier, and the Gammascope 101 was used with the Harshaw integrated crystal and photomultipler. Environmental samples have been readily measured with reproducible results. (REG)

Deep-Water Isotopic Current Analyzer*

Abstract: Three radicisotope-based instruments for determining current speed and direction and the early tests on them are described. The first two are radioactive tracer (^{131}I) drift instruments for use down to 400-ft and 1-mile depths, respectively. The third is a tilt meter, which has been tested at 15 ft, in which the angle of tilt is measured by the attenuation of beta radiation (^{106}Ru).

One of the many problems confronting both the hydrologist and the oceanographer has been the lack of a suitable method for precisely measuring and accurately recording the direction and speed of slow-moving currents, e.g., less than 2.5 ft/sec. In a program sponsored by the USAEC Division of Isotopes Development, three current velocity gages based on radioisotopes have been developed by Johnston Laboratories. The first two of these, designated DWICA I and II, are time-of-drift meters, designed for operation at depths found in lakes or rivers and in oceans—about 400 ft and 1 mile, respectively; DWICA III is a tilt meter and has been tested at 15 ft.

DWICA I and II

The DWICA I and II systems operate on the radioactive tracer principle. Tracer amounts of a radioisotope—^{131}I with a half-life of 8 days has been used so far—are injected into the stream under study. Radiation detectors, arranged in a circle of known diameter around the point of injection, record the passage of the tracer as it drifts past them. The time required for the tracer to reach a detector indicates the current speed, and the detector that responds first indicates the direction. Current speeds in the ultralow range are accurately measured, the theoretical lower limit being about 0.002 ft/sec, the rate of self-diffusion of the tracer. The upper limit is 2.5 ft/sec. Only the velocity component parallel to the bottom of the instrument is measured, and no inertial effects are shown since the tracer fluid has the same density as the stream water. The chief disadvantage of the instrument is the large weight in air of the underwater portion—240 lb for the latest model of No. I and 600 lb for the latest model of No. II—and the large cross-sectional area it presents to the stream which would cause errors in results obtained in small streams.

DWICA I

The DWICA I instrument used to demonstrate the feasibility of the system and the early tests have been described in detail elsewhere.[1,2] The DWICA IB, the model now being tested, is a modified version of the original. It consists of an above-water module housing the recording equipment and some of the electronics, which, during operation, is connected by a cable to an underwater module containing the measuring head (Fig. 1). The underwater unit, 81 cm diameter and

*Based on A Family of New Isotopic Water Current Meters (the DWICAS), Final Report, USAEC Report JLI-2748-86, Johnston Laboratories, Inc., 1966.

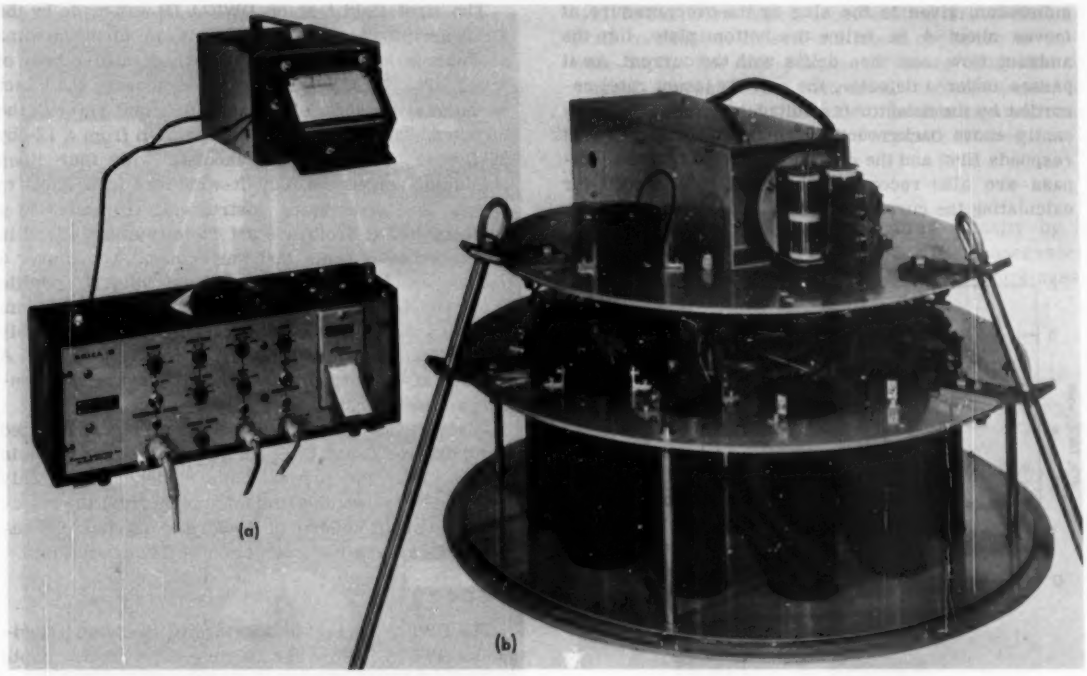

Fig. 1 The DWICA IB: (a) above-water unit; (b) underwater unit, supported on a tripod.

53.5 cm high, is constructed of three 50-cm-diameter decks of anodized aluminum, connected by stainless-steel rods. On the top deck is a 1200-ml rectangular reservoir containing the tracer solution—an aqueous iodide solution containing 5 mc of ^{131}I—and a differential-pressure module. Two canisters contain the power electronics and a compass with its readout electronics. There are 12 detectors on the bottom deck, equally spaced at the circumference of a 50-cm-diameter circle. Each detector consists of a scintillator crystal with a photomultiplier and preamplifier, sealed in an epoxy canister.

The apparatus used for injecting the tracer solution has several unusual features (Fig. 2). It is designed to give no appreciable horizontal momentum to the injected solution. And, since the force required for injection is provided by a pressure differential, it may operate at any depth regardless of ambient pressure. When an injection is started, a timing mark is recorded on the printout, and four constant-force springs act on a piston to apply a constant pressure of 6 psi above ambient water pressure to the solution in the reservoir. A volume of solution too small to affect the flow—0.2 to 2.0 ml—containing 2 to 12 μc of ^{131}I is discharged through an orifice at the center of the bottom plate as a toroid-shaped slug. The solenoid valve that closes this orifice has a variable opening time, from 0.1 to 0.6 sec. Because of the vertical

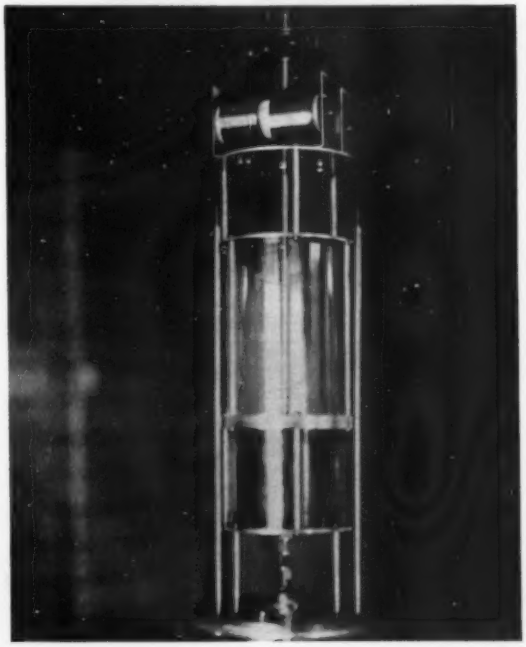

Fig. 2 Tracer solution injector.

momentum given to the slug by the overpressure, it moves about 4 in. below the bottom plate, into the ambient flow, and then drifts with the current. As it passes under a detector, the activity (count rate) recorded by the detector (as voltage) increases significantly above background (Fig. 3). The detector that responds first and the direction indicated by the compass are also recorded, giving all data needed for calculating the current velocity.

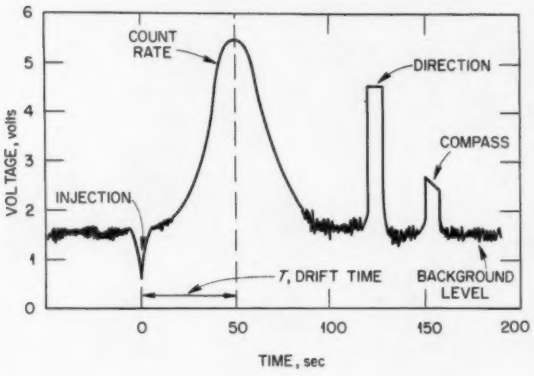

Fig. 3 Typical DWICA IB output. The drift time of 50 sec shows, from a calibration chart, a rate of 0.020 ft/sec. The direction reading of 4.5 volts indicates detector No. 9, and the compass reading of 2.5 volts indicates an orientation of 150° relative to magnetic north, or 270−150 = 120° clockwise from north.

The first field test on DWICA IB was made by the Tennessee Valley Authority (TVA) in North Carolina at Fontana Reservoir, a thermally stratified body of water. The TVA was interested in knowing the effect on current velocity of water withdrawal through the turbines. The instrument was operated from a 12- by 26-ft barge, which was moored with four lines stretching across the 2000-ft-wide lake from shore to shore. The underwater instrument, suspended to a depth of 350 ft from a steel cable, was stabilized in the water by a cable that was brought aboard over a sheave on the stern. A vertical velocity profile (Fig. 4*a*) obtained half a mile upstream from the dam indicates the instrument's capability of accurately measuring slow velocities (from 0 to 0.15 ft/sec). A horizontal velocity profile (Fig. 4*b*) in the same region shows the extent of flow uniformity.

A river version of DWICA IB has been constructed and delivered to the U. S. Public Health Laboratory in Athens, Ga., for testing. This model contains only three sensors, which are downstream from the injector. The small number of sensors is satisfactory because the approximate direction of flow is known.

DWICA II

The DWICA II (Fig. 5) operates on the same principle as DWICA I. However, because of the greater depth at which DWICA II will be used, corrosion and pressure in the underwater unit are more serious problems. Corrosion was virtually eliminated by making the small parts of stainless steel and coating the large aluminum parts with an epoxy compound. For

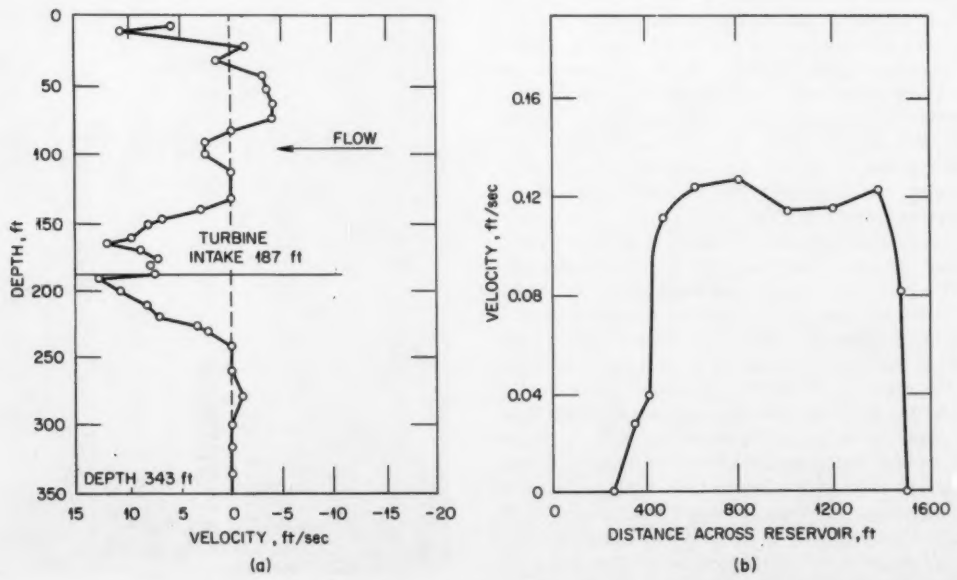

Fig. 4 Velocity profiles, (a) vertical and (b) horizontal, in Fontana Reservoir.

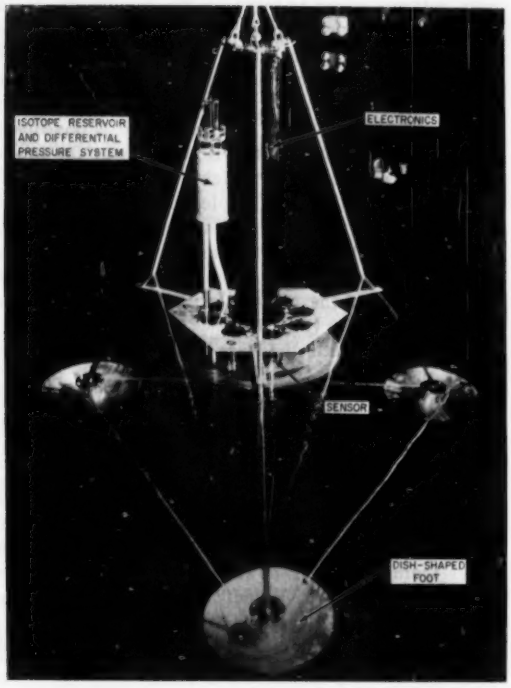

Fig. 5 The DWICA II.

pressure protection, the electronic circuitry and sensors were enclosed in pressure cases.

There are only 8 sensors in DWICA II, but a special logic circuit gives an angular determination of direction equivalent to that of 32 sensors, and directions are indicated within ±12°. A horizontal baffle plate under the sensor pressure cases ensures laminar flow of the stream past the detectors. To make the end caps on the sensor pressure cases thin enough for tracer amounts of radioactivity to be detected through them but strong enough for deep-water pressures to be resisted is very difficult. However, satisfactory caps were made from an aluminum alloy by Ocean Research Equipment, Inc., who developed a new electron-beam technique for welding this material.

The components of the underwater unit are mounted on a tripod, which is an integral part of the unit. In the latest modification the tripod legs have dish-shaped feet, which can hold the structure in a vertical position in most ocean-bottom undertows. A computer in this unit programs the frequency and size of the tracer injections, depending on the current speed.

Numerous underwater tests, made in cooperation with the Navy, have proved the performance of the DWICA II system. It has operated successfully at 140 ft and has accurately measured flow rates of 0.17 to 1.67 ft/sec. Now under consideration are modifications to adapt it to self-sufficient operation at a 1-mile depth. For this purpose buoyancy equipment to control descent of the instrument and an expendable bottom anchor would be added. Remote control from shipboard would be effected by acoustic devices.

DWICA III

The DWICA III system measures velocity by a radiation-absorptiometry method. The underwater unit (Fig. 6) consists of a cylindrical pressure case,

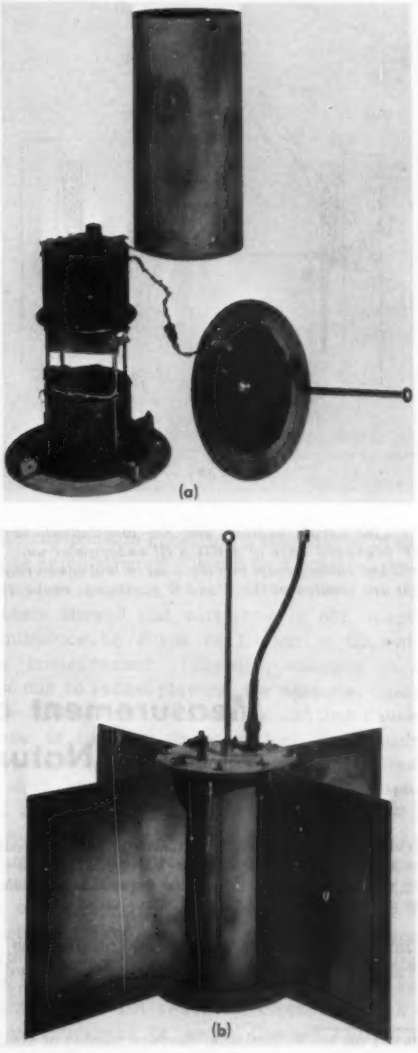

Fig. 6 (a) Components and (b) assembly of DWICA III underwater unit.

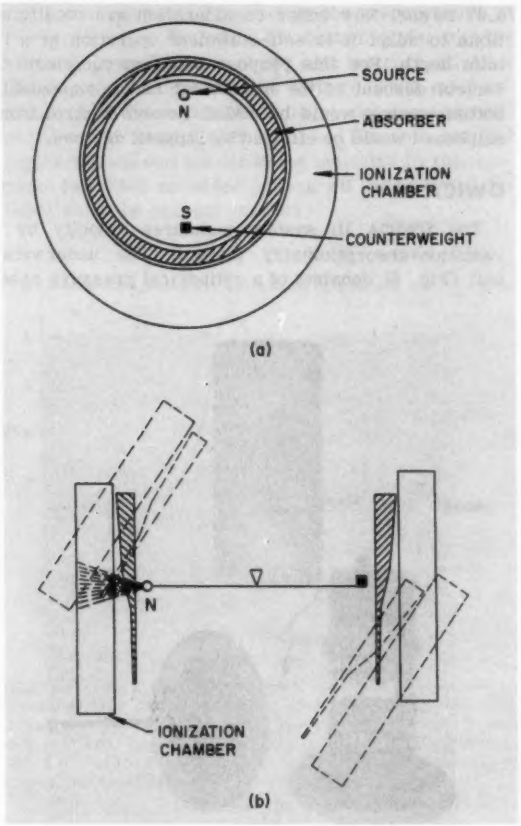

Fig. 7 (a) Cross section and (b) longitudinal section of upper pressure case of DWICA III underwater unit. Lower case is the same except that the source and counterbalancing weight are located at the E and W positions, respectively.

with optional vanes to increase the sensitivity. Inside the case are two cylindrical ionization chambers, each of which surrounds a Lucite absorber cylinder with tapering walls. A compass disk mounted on a pivot inside each absorber cylinder has a beta-emitting source, 400 μc of ^{106}Ru, located at the N position in the upper assembly (at the E position in the lower) and counterbalanced by a weight (Fig. 7*a*). As the current speed changes, the whole unit tilts at an angle, the square root of whose tangent is proportional to the speed. The angle of tilt is determined from measurements of the current in the ionization chambers, which is a function of the amount of radiation transmitted through the Lucite (Fig. 7*b*).

The North–South and East–West velocities determined in the two sections of the underwater unit are transmitted to the above-water unit, which has two separate print-out channels. The two vectors are then averaged for a selected time interval of from 0.25 to 6 hr.

The Chesapeake Bay Institute used the DWICA III prototype, unattended, to measure flow patterns in Lake Ontario in connection with the proposed waste outlet from a nuclear power plant to be constructed near Rochester, N. Y. Velocities as low as 0.08 ft/sec were successfully measured at a depth of 15 ft over a 2-week period. (Martha Gerrard)

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Measurement of Snow-Water Reserves Using Natural Soil Radioactivity*

Abstract: The results of preliminary tests indicate that under some conditions it may be feasible to measure snow-water reserves by determining the degree to which the snow cover attenuates the natural soil radioactivity.

An interesting method for determining the water equivalent of the snow cover has been proposed by the USSR Hydrogeological Institute. Although it would undoubtedly have limited application, the results of preliminary tests have shown good correlation with those from standard snow-withdrawal methods† under the condition tried.

*Based on N. V. Zotimov, Surface Method of Measuring Snow-Water Reserve Using Soil Radioactivity (in Russian), *Trudy Gosudarstvennogo Gidrologicheskogo Instituta*, 130: 148-162 (1965).

†Standard methods of snow-water measurement, including the use of radioisotope gages, were reviewed by J. L. Smith and D. W. Willen, Radio-Snow Gages: A Review of the Literature, *Isotopes and Radiation Technology*, 2: 41-49 (1964); and by J. L. Smith, Instrumentation for Snow Gaging—Yesterday, Today, and Tomorrow, *Isotopes and Radiation Technology*, 4: 227-237 (1967).

The method is simple in principle: The natural radioactivity at the surface of the earth—due chiefly to ^{40}K and the uranium and thorium series of nuclides—is measured in the absence of a snow cover and is then measured again, at the surface of the snow, after a snowfall. The activities so measured, N_0 and N (counts/min), respectively, are related to the mass absorption coefficient of the snow, μ, and to the thickness (and density) of the absorbing layer, h (g/cm^2), by

$$N = N_0 e^{-\mu h} \quad (1)$$

Since the absorber is actually the snow cover plus the layer of air between the surface of the snow and the detector, for h we substitute the term $S + h_{air}$, where S is the millimeters of water represented by the snow layer and h_{air} is the thickness of the air layer. Then we can find S directly in millimeters of water reserve from logarithmic expression of Eq. 1,

$$S = (\ln N_0 - \ln N)/\mu - h_{air} \quad (2)$$

For the Hydrogeological Institute tests, a nomogram was constructed and calibrated so that S could be read directly (Fig. 1). Under the conditions of the test, for $N_0 = 1500$ and $N = 1150$ counts/min, $S = 81$ mm of water, for example. The agreement with values found by standard methods at two different points on two different days was good:

Date	Natural radioactivity method	Snow-withdrawal method
12/29/63	35	36
	40	40
1/30/64	102	90
	82	88

In applying formula 2 the value of h_{air} may be neglected if the detector is only 1 to 2 m above the surface of the snow, e.g., in a truck. The absorption of gamma radiation by a 1-m layer of air is about the same as that of snow equivalent to 1.3 mm of water. However, h_{air} becomes equivalent to 32 to 64 mm of water when the detector is 25 to 50 m above the earth and therefore must be included if the detector is airborne.

The value of μ was determined by measuring the earth's background simultaneously with the snow-water reserves by a snow-withdrawal method. It was found to be

$$\mu = 0.053\, S^{-0.223}\ \text{cm}^{-1} \quad (3)$$

Because of the wide gamma energy spectrum—0.5 to 3.0 Mev—of the natural radiation, μ decreases with increasing absorber thickness and only the more energetic components penetrate a thick layer of snow. Therefore calculation of μ would be a complex procedure.

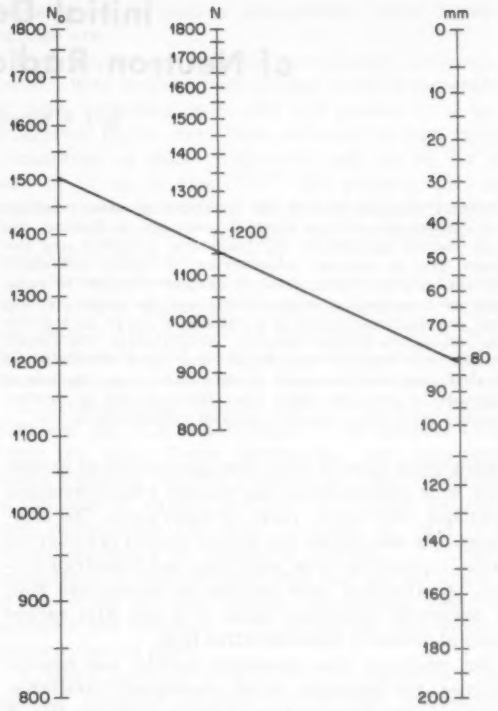


Fig. 1 Nomogram, calibrated for field radiometric equipment, used for determining snow-water reserves by natural radioactivity method.

The tests showed that variations in soil moisture could introduce an error of 10 mm in the water-reserve measurement. Likewise, changes in soil porosity due to recent plowing, for example, changed the soil–atmosphere air exchange and thus caused a difference in the soil radon content. Variations in groundwater levels, if near the surface, affected N_0 values. And, of course, since the mineral content of the soil differs from place to place, if N_0 and N are not measured at the same points, the results are not consistent. Other sources of error include differences in the rate of detector movement, differences in the number of detectors in the detector unit, and the distance between measurement points. Beta radiation is eliminated from N_0 measurements by using a suitable filter or a counter that responds only to gamma radiation, and it does not reach the counter through any significant thickness of snow. Cosmic radiation is considered not to differ enough between the N_0 and N determinations to affect the results significantly.

(Martha Gerrard)

Initial Development of Neutron Radiography in France*

By J. P. Barton† and J. L. Boutaine‡

Abstract: Development of the technique of using neutrons for radiography started about 2 years ago in France, and both routine application and long-term research are now under way in several laboratories. A highly collimated thermal-neutron beam from a nuclear reactor is being used as a method of examination, and the technique may find valuable application in a variety of fields both inside and outside the nuclear industry. Nevertheless, the subject is new and largely unexplored, and more sophisticated methods may be developed in the future; e.g., the use of neutrons of energies other than thermal and of neutron sources other than reactors, such as radioisotopes.

Radiography using X rays from generators or gamma rays from radioisotopes has become a well-developed technique over many years of experience. The techniques are well known and widely applied in numerous fields — research work, medicine, and industrial control. Applications have become so widespread that, in industrial countries, sales of X-ray film exceed those of ordinary light-sensitive film.

But neutrons also penetrate matter, and neutron sources are becoming more conveniently available. What are the possibilities of using neutrons like X rays? And what might be the advantages? Some studies of these questions were started recently in France. Before 1960 only three research papers[1-3] had been published on neutron radiography — a contrast to the many volumes written on the development and refinement of X-ray techniques. The first two publications described the early work done in Berlin in 1935 to 1939, following Chadwick's[4] observation in 1932 of the penetrating nature of neutron radiation. The third was a journal publication of an earlier British report. The first known routine applications were at Argonne National Laboratory[5] in 1962 to 1964, and about this same time work was resumed in both Britain[6] and Japan.[7,8] An extensively documented book[9] on neutron radiography published in 1965 includes detailed reviews of procedures and related subjects that are outside the scope of this review, and interest in the technique is steadily increasing.[10]

In France work started in 1965, and some simple procedures introduced in the last few years at the Centres d'Etudes Nucléaires (CEN) at Grenoble, Cadarache, and Saclay are proving useful in routine application. For example, a principal objective of the research reactors is to study the effects of irradiation on a variety of objects. Sometimes these are experimental or operational reactor fuel elements; sometimes they are other materials irradiated perhaps under controlled stress or temperature. Neutron radiography of these objects at intervals during their irradiation, or at the end of their irradiation, is a method of examination now proving both effective and convenient.

In addition to some varied interests within the Commissariat à l'Energie Atomique (CEA), several other industries in France are now actively involved in this field, and a general invitation is extended to all industries to suggest new applications. It is hoped that through research and development the possibilities will be more widely explored.

Advantages

Probably the most far-reaching possibilities of neutron radiography arise from the fact that the information obtained by it is fundamentally different from that obtained with X or gamma radiography. In some cases details are revealed by neutron radiography which cannot be obtained by other means of examination, so that neutron radiography is not competitive with but complementary to X radiography.

This advantage is due to the basic differences between the interaction of neutrons and of X rays with matter — neutrons interact with the nucleus, while X rays interact with the orbital electrons of an atom. Thus absorption of X rays of normal penetrating energy by a material increases with the atomic number of the elements making up the material. But the nuclear properties of elements do not change in this regular way, and the absorption coefficient of matter for neutrons may change abruptly between one element and its neighbor in the periodic table. This change may be by several orders of magnitude (Fig. 1). Materials with low neutron attenuation, such as aluminum, lead, and uranium, are easily penetrated (left-hand ordinate), while those with high neutron attenuation — boron, cadmium, gadolinium — provide excellent contrast (right-hand ordinate), and very small thicknesses can be distinguished. The differences between the two types of radiography are clearly illustrated in Fig. 2. Some

*This is the original English paper from which the French version in *Supplement au Bulletin d'Information de A.T.E.N.*, No. 66, pp. 4-9 (July–August 1967), was prepared.

†Formerly visiting scientist at Grenoble University, working in association with the Centre d'Etudes Nucléaires; present address: Argonne National Laboratory, USA.

‡Section d'Application des Radioéléments, Centre d'Etudes Nucléaires, Saclay, France.

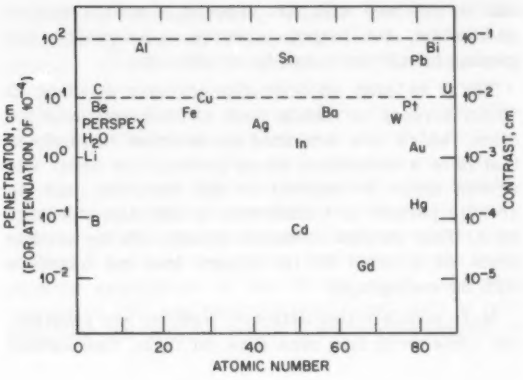


Fig. 1 Penetration of various elements by thermal neutrons and contrast shown by these elements.

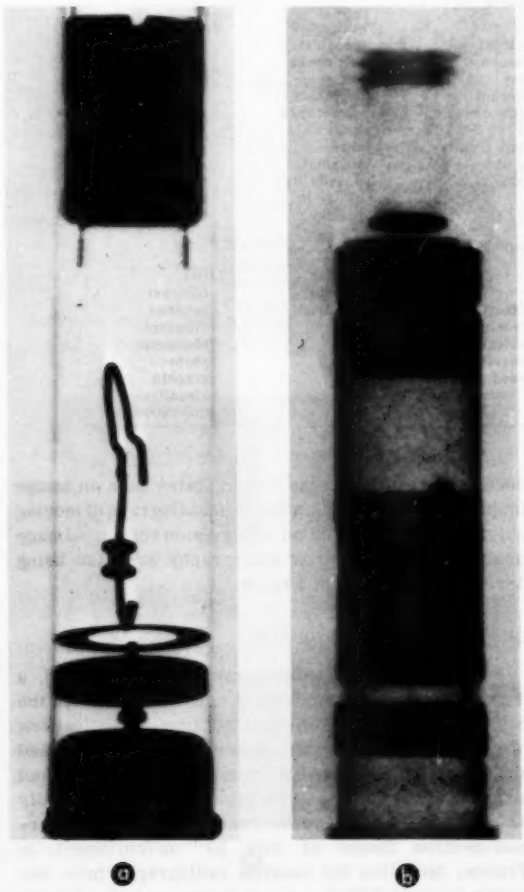


Fig. 2 Different structures revealed by (a) an X radiograph and (b) by a thermal-neutron radiograph of the same object.

cases in which neutron radiography shows an advantage are:

1. *Observation of light materials behind heavy materials:* With neutron radiography, small thicknesses of light materials, e.g., 100 μ of plastic or of hydrogenous liquid, have been revealed through large thicknesses of dense materials such as 30 cm of lead or 10 cm of steel.[6,11,12] This property may be applied in examination of electrical insulation, gaskets, or liquids such as water, oil, or gasoline enclosed in dense metal.

2. *Distinguishing between materials of similar atomic weight:* Materials of neighboring atomic weight and density are difficult to distinguish with X rays but often show good contrast in neutron radiography. Examples are tin and silver, lead and mercury.

3. *Distinguishing between isotopes of an element:* Although the different isotopes of an element have about the same X-ray attenuation, they may have vastly different neutron attenuation. This difference in the case of certain isotopes is so great that neutron radiography has been proposed as part of a tracer technique in biological studies.[13–15] Another application in this category that has proved useful[16] is the study of ^{113}Cd burnup; this isotope captures neutrons and becomes ^{114}Cd during use in a nuclear reactor control plate.

4. *Observation of radioactive materials:* The gamma radiation emitted by strongly radioactive objects can interfere with an X radiograph, but neutron radiographs may be made by methods that are insensitive to this interfering radiation. Radioactive fuel elements from a nuclear reactor are now being examined[5,17,18] by this new technique.

5. *Convenience:* In some cases neutron radiography may actually be more convenient than X-ray inspection. For example, in the examination of objects near a pool reactor. A strong source of neutrons is already present, the apparatus required is cheap and simple, and radioactive objects may be manipulated easily below the shielding water, thus avoiding the usual handling difficulties.[17,18] In some other cases, neutrons may be more powerful than X rays for penetrating thick sections of dense metals, such as iron, where there is a significant buildup of scattered X radiation. Examination of the uranium and lead used for radiation shielding has been suggested as an application for neutron radiography.[19]

Techniques

CHOICE OF NEUTRON ENERGY

It has been demonstrated experimentally that radiography may be performed with neutrons of four distinct energy ranges:

1. Fast neutrons directly from a neutron source[20]
2. Resonance-energy neutrons of specifically selected energy[21]

3. Thermal neutrons directly from a moderator surrounding the source[1-3]

4. Cold neutrons, of energy less than normal thermal energies[22]

The simplest technique is that using thermal neutrons, and almost all neutron radiography work has been concentrated in this energy range in the past. Neutrons of normal thermal energies are the most easily obtained, and it is a fortunate coincidence that within this energy range an interesting series of contrasts can be obtained. However, for certain applications, other neutron energies might show advantages. The characteristics of these different types of neutrons for radiography are shown in Table 1. Very little work has been devoted to the development of techniques using fast, resonance, or cold neutrons, and further study in these directions is indicated.

Table 1 COMPLEMENTARY RADIOGRAPHY TECHNIQUES

X rays	Gamma rays	Cold neutrons (< 0.005 ev)	Thermal neutrons (~0.025 ev)	Resonance neutrons (0.1–10 ev)	Fast neutrons (kev–Mev)
			Advantages		
Good-quality radiographs with small generators; possibility of varying voltage and using filters	Small-size sources; high penetrating power	High penetration of steel; little diffusion; high contrast for hydrogenous materials	High contrast between many different elements; easy to obtain from a neutron beam	Very high penetrating power for some elements; sometimes better than thermal; possibility of use in biology	Very high penetrating power; possibility of using neutron generators as point sources (no thermalization or loss by collimation)
			Limitations		
Limited sensitivity; dense materials more absorbent than light ones		Research reactor required	For thick objects, sharpness lacking unless reactor used	Research reactor required	Little contrast between different elements; photographic visualization difficult

METHODS OF PHOTOGRAPHIC DETECTION

Films are relatively insensitive to neutrons, and the most convenient method of recording neutron radiographs is to place a special converter screen next to the film. The screen captures the neutrons and emits either alpha or beta particles or gamma rays, which in turn affect the film. Another very useful technique is to mix the neutron-capturing element with a scintillator and use this next to a light-sensitive film. The best such systems provide a picture with very low neutron exposure (below 10^5 n/cm^2) and at the same time have excellent discrimination against gamma radiation. The method is therefore very important for radiography with small neutron sources. Better-quality neutron radiographs can be obtained with thin screens of metals such as gadolinium, but longer exposure is necessary, and gamma interference may be troublesome.[23]

There is also an activation-transfer process, in which screens of metals such as dysprosium or indium, which are activated by neutrons to products that have a convenient decay period, are used. The screen alone is exposed to the neutrons, and the activity pattern is transferred to the film at a later time. Thus gamma radiation mixed with the neutron beam or emitted by the object does not interfere with the radiograph.

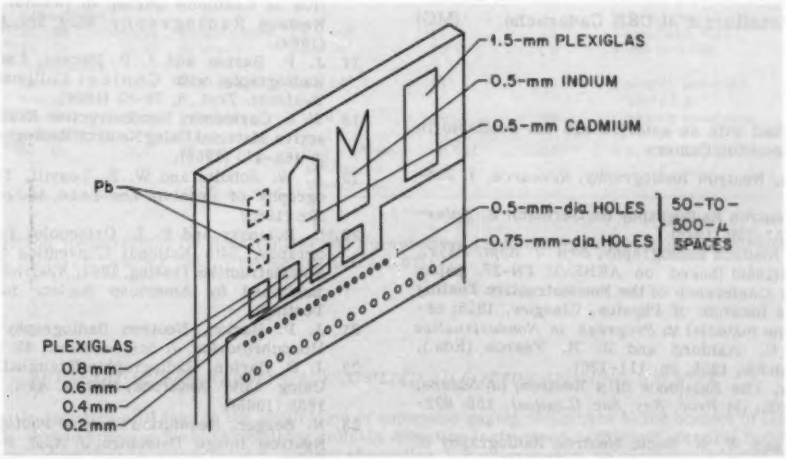
More sophisticated detection methods are possible, but little work has been done on them. One method developed recently in the United States uses an image intensifier and enables a neutron radiograph of moving objects to be displayed on a television screen.[24] Image intensifiers for neutron radiography are also being developed by a firm in France.[10]

CHOICE OF NEUTRON SOURCE

For high-quality neutron radiographs (Fig. 3), a thermal-neutron beam from a nuclear reactor is the best source of neutrons, and all useful applications of neutron radiography thus far reported have used such beams. A high-flux research reactor is not absolutely necessary, and good quality can usually be obtained with low-power reactors, providing thermal-neutron fluxes of only 10^{11} n/(cm^2)(sec). In France, facilities for neutron radiography have recently been installed, or are in the process of being installed, on the Melusine, Siloé, Pegasus, Peggy, Isis, and Osiris reactors.

However, although many objects that are to be examined by neutron radiography can be taken to a nuclear reactor, the usefulness of the technique certainly would increase if other neutron sources could be used. Studies are now under way at Grenoble on the possibility of using small accelerator neutron sources, similar in size and cost to X-ray sets.[25] Results to date show that thermal-neutron radiographs of moderate quality can be obtained with small, inexpensive neutron generators and direct-exposure techniques. It has also been shown that the neutrons from existing generators of the 400-kv class, and with outputs of 10^{11} fast neutrons per second, may be just sufficient for making useful neutron radiographs of radioactive objects. The possibility of performing neutron radiography in high-activity handling cells using a neutron generator is receiving serious consideration. Such a procedure is of interest to a number of CEA, Electricité de France, and Euratom laboratories.

Some work has also been done in France on the possibility of using radioactive neutron sources. The conclusion at present is that one particular radioisotope source of neutrons could be useful for certain applications of neutron radiography. This is ^{241}Am–Be, sources of suitable strength having recently become available. Americium, which has a half-life of 458 years, is produced by long irradiation of



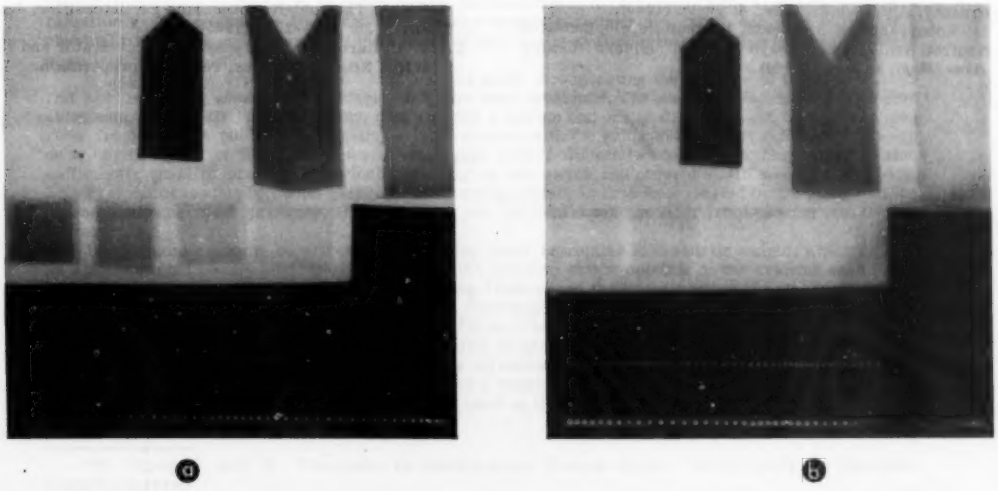


Fig. 3 Quality of neutron radiographs made by (a) neutrons from a nuclear reactor and (b) 14-Mev neutrons from a generator, 4×10^{10} n/(cm^2)(sec). Lead is transparent to the neutrons and therefore does not show.

plutonium in high-flux reactors and can be separated from spent fuel elements. Radiographs of moderate but useful quality can be obtained by the direct-exposure technique, the low source strength being compensated by long exposure times. Small sources would be reasonably portable and relatively inexpensive if rented from a central agency when needed.

Within the CEA, research workers in various groups are contributing to the development of the subject. These groups are La Section d'Application des Radioéléments at CEN Grenoble and CEN Saclay, La Section Physique Experimentation du Département des Piles Expérimentales at CEN Saclay and CEN Cadarache; Le Service des Piles and La Section des Accélérateurs at CEN Grenoble; and Le Groupe d'Etude des Laboratoires de Haute Activité du Département de Metallurgie at CEN Cadarache. (MG)

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NEUTRON RADIOGRAPHY CONFERENCE

A 1-day information meeting* on neutron radiography was held at Saclay, France, in October 1966. Five papers were presented, which included, in general, the same information as that given in the above review by Barton and Boutaine but in considerably greater detail. Part of a tabulation of radioisotopic neutron sources from the conference proceedings is reproduced here in modified form because of its special interest to persons in the radioisotope field:

Radioisotope Neutron Sources

Source	Reaction	Half-life	Emission from 1 curie, fast n/sec	Maximum neutron energy, Mev	Background, mr/hr, for 10^6 n/sec at 1 m	Gamma energy, Mev
^{241}Am–Be	(α,n)	458 years	2.2×10^6	11	0.1	0.060
^{210}Po–Be	(α,n)	138 days	2.5×10^6	11	0.04	0.79
^{242}Cm–Be + daughters	(α,n)	167 days	2.5×10^6	12	10	Fission-product spectrum
^{239}Pu–Be	(α,n)	24,400 years	1.8×10^6	11	1.7	Essentially 4.45
^{227}Ac–Be + daughters	(α,n)	22 years	1.5×10^7	13	8	0.009 to 0.190
^{226}Ra–Be + daughters	(α,n)	1620 years	1.5×10^7	13	60	Complex spectrum up to 1.8
^{228}Th–Be + daughters	(γ,n)	1.91 years	2×10^7	14	30	Complex spectrum
^{124}Sb–Be	(γ,n)	60 days	1 to 5×10^6 depending on dimensions	0.024	1000	0.6, 0.7, 1.7, 2.1

*Neutron Radiography: Its Possibilities and Perspectives (in French), Proceedings of Information Meeting, Saclay, 5 October 1966, CEA Report DR/AR/67.1.

(MG)

*BETA-RAY SLIVER WEIGHT GAGE APPLIED TO COTTON SPINNING MACHINE**

The textile industry still lags in the application of automatic gaging techniques to the control of cotton sliver-to-thread spinning because obtaining reliable detecting instruments is difficult. Several methods of gaging—capacitance measurement, optical absorption, air micrometer, or mechanical feeler such as the "Autoleveller"—have been developed for on-line measurement and control, but these have not proved successful in practice. In recent years, the advantages to be gained by the application of beta-radiation gaging techniques to sliver weight measurement have been discussed, but, despite the basic physical possibility, no commercial instrument of this type is available that satisfies the requirements of reliability, cost, and safety.

In Japan a new type of gage in which a sliver guide incorporating the windows of the ion chamber and the radiation source as a single unit has been developed. The sliver guide, made of 0.03-mm aluminum, has an air jet at the entry, which guides a broken end of the sliver into the hole. The sliver runs freely in the guide hole, making a loose contact with the guide walls—just sufficient to sweep away any flying cotton fiber, which would cause gage drift if allowed to accumulate. The contact is also sufficiently good to provide electrical grounding of the sliver and prevent static electricity buildup. No error is caused by position change of the running sliver, as it is well located by the guide and the detector arrangement prevents any radiation not incident upon the fiber from passing into the ion chamber.

The dynamic weight of sliver while running must be related to the static weight, which is used as a conventional quality control of sliver since the dynamic weight depends on the drawing conditions. Good correlation between the effect of drawing tension and the appearance of the running sliver in the guide was obtained with the use of a constant determined by the drawing speed.

The sliver weight gage uses 15 mc of ^{204}Tl as a source and an all-solid-state electrometer as a detector. A minimum range of 100 grains ±10% is provided (100 grains = 6.48 g). In actual use on the drawing machine, the dynamic weight gives an accuracy within ±0.2% for 400 grains. Tests have been made for both continuous operation of over a month's time and use immediately after switching on. The beta-ray sliver weight gage will be used as the detector unit of a cotton spinning machine controller.

(FJM)

*M. Katagiri and H. Takahara, in *Radioisotopes (Tokyo)*, 16(11): 11-16 (1967) (in Japanese, English abstract).

Section

III

Isotopes and Radiation Technology

Food Irradiation

Seventh Annual AEC Food-Irradiation Contractors' Meeting

By F. E. McKinney

Abstract: This article summarizes the brief reports presented by the USAEC food-irradiation contractors at their seventh annual meeting in Washington, D. C., on Sept. 27, 1967. Division of Isotopes Development contractors gave 19 reports, and Division of Biology and Medicine contractors gave 21 reports. Several products are lacking only the Food and Drug Administration approval to become commercialized.

On Sept. 27–28, 1967, the seventh annual meeting of the food-irradiation contractors of the U. S. Atomic Energy Commission (AEC), of both the Division of Isotopes Development (DID) and the Division of Biology and Medicine (DBM), was held in Washington, D. C. The first day was spent in four technical sessions on (1) fruits and vegetables, (2) marine products, (3) microbiology, and (4) wholesomeness. On the second day, summary reports were presented by all contractors. The program was co-chaired by Dr. Kevin Shea of DID and Dr. William Pace of DBM. After opening remarks by the chairmen and by Eugene Fowler, Director of DID, and Dr. John Totter, Director of DBM, the contractors presented the following results.

DID Contractors' Reports

P. Markakis, Michigan State University, reported on the irradiation of fruits and vegetables — especially apple juice, asparagus, mushrooms, and strawberries. A dose of 10 to 100 krads delays opening of caps of mushrooms very effectively. Mushrooms are not a staple, nor are they a large-volume sales item; but they are expensive, and radiation costs could be absorbed easily. A calcium treatment used on sweet cherries, strawberries, and blueberries prevented softening of fruit after irradiation.

E. M. Ahmed, University of Florida, gave results of combining radiation with a hot water dip in treatment of citrus fruits. The same extension of shelf life could be achieved with half the dose. Peel injury was shown to be dependent on physiology of fruit, dose rate, and storage conditions. The hot water dip had no effect on the ascorbic acid and sugar in fruit. The fruit had more juice after the combination treatment than after the radiation treatment alone, but this is due to the lowered dose.

E. Ross, University of Hawaii, described the operation of the University Experiment Station. Technical work has shown that the respiration activity of five tropical fruits is directly correlated with the radiation dose. The avocado is the least radiation tolerant of all fruit studied. Some work has been conducted to determine at what dose irreversible changes in the fruit occur.

The Director of the Hawaii Development Irradiator (HDI), K. K. Otagaki, reported that the HDI was completed in June 1967 and dedicated in July 1967. The goal of operation is the commercialization of the radiation process for tropical fruits, with emphasis on disinfestation. Cost studies have been encouraging. Dr. Otagaki says that current operating costs are $70/hr using four plaques. Shipping studies with irradiated papayas have been performed with the full cooperation of commercial carriers. Commercialization of the process within 2 to 3 years is hoped for.

J. Cuevas, Puerto Rico Nuclear Center, presented results of irradiation of plantains, mangoes, and bananas. A survey of the mango market potential in the United States was initiated. Irradiation of bananas is being curtailed as a result of cost-benefit studies. Shipping studies with irradiated mangoes involving airfreight between San Juan and Miami are in progress.

The progress in the fruit and vegetable area was summarized by Dr. Shea. E. C. Maxie, University of California, pointed out that strawberries are still the most promising fruit for commercialization.

Markakis emphasized the importance of the new calcium treatment to prevent irradiation-induced softening. Ahmed said that peel injury in citrus fruit, while displeasing to the eye and to the consumer, does not affect the quality of the fruit. The U. S. Department of Agriculture has shown strong interest in irradiating papayas for fruit-fly disinfestation and mangoes for seed-weevil control.

The next series of reports covered progress in radiation treatment of fish and shellfish. The first report was given by L. J. Ronsivalli, Bureau of Commercial Fisheries, Gloucester. Work has been done to determine the optimum dose for radiation pasteurization of Northwest Atlantic finfish and to establish maximum edible shelf life. Work to lower the dose has so far shown that the dose specified in the petition to the Food and Drug Administration (FDA), 100 to 200 krads, is the lowest effective dose.

John Kaylor, also of the Bureau of Commercial Fisheries, described operation of the Marine Products Development Irradiator (MPDI) during the past year. Over 100 irradiations of over 40 thousand lb of product have been done. Irradiation of cod and haddock fillets has given good results, and shipping studies have shown that fish purchased on open markets can be irradiated and shipped from the East Coast to Washington State and still be in marketable condition. A dose of 200 krads will double shelf life of oysters. Free liquor* from oysters remains a problem, but this may not be a barrier to commercialization since the regulation on allowable amount of free liquor is set by the U. S. Public Health Service.

M. R. Rao, Louisiana State University, gave results of the study of proteolytic action in irradiated shellfish. A slight decrease in nutritive value occurs during ice storage of oysters; texture changes were also noted. There may be a significant correlation between the decrease and the texture changes. Organoleptic studies at Ft. Lee, Va., have consistently shown that the irradiated product is preferred over the nonirradiated control.

At the morning coffee break, irradiated Hawaiian papayas were on display.

D. Miyauchi, Bureau of Commercial Fisheries, Seattle, reported on trucking studies of irradiated Pacific Coast finfish. Over 2000 lb was shipped in 14 shipments to Chicago, Kansas City, Denver, and San Diego. Refrigerated trucks were used, and the fish were stored in commercial cold-storage rooms after reaching their destination. Industry evaluations termed the irradiated product excellent. Chemical studies show that glucose content can be used as a quality index and that ribose content cannot. The glucose/ribose ratio may be useful to indicate that a product has been irradiated, but this possibility must be confirmed with additional studies.

Freshwater fish have been radiation pasteurized by the Bureau of Commercial Fisheries at Ann Arbor, Mich., reported John Graikoski. Perch fillets were irradiated and stored in an atmosphere of low oxygen content. Fillets inoculated with 10^8 organisms per gram and stored in air at 35°F spoiled in 18 days; in a controlled atmosphere under the same conditions, the fish did not spoil for 24 days. The effect of irradiation on yeast and mold growth on the product was quite pronounced. He noted that they are having the usual difficulties with the difference in acceptance of the irradiated product by expert and amateur panels.

M. S. Silberstein, IRRADCO, described the proposed Pilot Plant Meat Irradiator (PPMI). IRRADCO is the official new name of Irradiated Food Products Co., which is a composite company made up of Allen Products Co.; Martin–Marietta; Isotopes, Inc.; Uniroyal; and a subcontractor, Thomson Farms. The PPMI will be located in Allentown, Pa. The AEC is financing the initial ^{60}Co source, and the Department of Defense is contracting for 300 thousand lb of irradiated ham, when the ham petition receives FDA clearance. IRRADCO expects also to irradiate 400 thousand lb for Allen Products Co. and 300 thousand lb for exploratory studies by other companies.

C. Wootten, Isotopes, Inc., announced the operation of the portable ^{137}Cs irradiator. Work in FY-1967 concentrated on training and paperwork. The operator had to develop test procedures that were standard but that could be modified for the different tests across the country. The unit is now operating in Arlington, Mass. It made a stop in Westport, Pa., to irradiate mushrooms.

F. X. Rizzo, Brookhaven National Laboratory (BNL), gave results of BNL studies on facility design and dosimetry. A new dosimeter to cover the range 3×10^5 to 3×10^6 rads has been developed. Flow dosimetry for such large systems as continuous grain irradiators is under study. He announced that there will be an international workshop on dosimetry in March at the National Bureau of Standards (NBS). Designs are being set up for the irradiators to be built for loan to Argentina and India.

W. L. McLaughlin, NBS, also spoke on dosimetry studies. He has developed paper dosimeters and was willing to give samples to anyone for testing.

The lack of FDA approval was termed the greatest barrier to industry acceptance of radiation processing by D. M. Yates, Battelle Memorial Institute (BMI). Operating under a subcontract from AEC funding to the Business and Defense Services Administration, BMI workers have made a commercial study of several foods. The outlook on commercialization of radiation processing of fish is promising, for de-

*Free liquor is a clear exudate of raw oysters.

hydrated vegetables excellent, for bananas uncertain, and for poultry good. Investment for an isotope plant was estimated at $3 million, with cryogenic facilities costing even more. It may be 20 years before radiation processing of food is routine.

L. E. Brownell, Battelle–Northwest Laboratories, reported studies using decaying reactor fuel elements. Potatoes were studied for effects other than inhibition of sprouting; duplication of growing 2 crops in 1 year as reported by the Russians was attempted successfully. Although the overall average yield per plant was smaller, the actual yield for individual plants was either nothing or vastly increased.

E. A. Garlock, Hazleton Laboratories, gave current status of information for petitions. The orange petition was withdrawn at FDA request; it will be resubmitted with more information. The strawberry petition has also been withdrawn due to lack of 2-year feeding studies. The petition was based on data from 90-day animal feeding studies and supported by previously reported wholesomeness studies on other related irradiated foods. The FDA has now determined that certain radiation-processed foods should be subjected to the more exhaustive 2-year tests. The U. S. Army Natick Laboratories has done some induced-activity studies in support of petitions.

E. L. Mayer, Savannah Grain Irradiator, presented results of studying stored product insects. Various levels of radiation were used on insects in various stages of development. A dose of 20 to 25 krads will be sufficient to sterilize beetles but not moths. For most species, females are more resistant to radiation than males.

W. Urbain, Michigan State University, summarized the cost-benefit studies on red meats of Daniel Yankelovich, Inc. He also presented results of experiments to retain red color and control of drip in irradiated meats. Net tangible benefits for commercialization of radiation processing of meats for centralized meat cutting will probably accrue in largest amounts when the process is used by chain stores. Technical problems include microbiology, color changes, and exudate. Work so far shows that phosphate treatment will control exudate, and controlled atmosphere the color change. The preferred dose is 50 to 100 krads. There seem to be no technical difficulties.

DBM Contractors' Reports

P. Appel, University of California at Berkeley, reported on radiation of heme proteins and the changes in structure of myoglobin subjected to gamma irradiation at 500 krads; peptide cleavage occurs at the C terminal end of the myoglobin. Extent of peptide cleavage is quite *p*H dependent, and control of *p*H may be a way to control damage to myoglobin by gamma radiation.

W. W. Nawar, University of Massachusetts, gave the results of a study of radiation effect on lipids. Mackerel that were at the laboratory within 2 hr of being caught were used as a source of oil. All irradiation was done under vacuum. The study has identified 53 components formed in fish lipids by radiation. These researchers have also developed tables that aid in the deduction of flavor threshold levels of the compounds.

L. Massey, Cornell University, presented the results of fundamental research on texture and biochemical changes in fruit aimed at preventing softening after irradiation. Apples stored for long periods after receiving 50 krads soften at a much slower rate than do nonirradiated ones. At the end of 60 days, they are the same firmness as the controls; at the end of 120 days, they are much firmer. These researchers also examined the problem of cell membrane permeability. High doses of radiation produced the same changes as freezing would. The relation of calcium leaching to firmness was noted. A glyoxylate pathway in carrots, lettuce, and apples has been discovered that may account for the CO_2 in the irradiated product.

F. J. King, Bureau of Commercial Fisheries, reported results of attempts to correlate odor of fish with irradiation. Over 30 compounds have been identified from irradiated clams; apparently dimethylsulfide is the predominant compound that is produced in irradiated clams. For haddock, no compound predominated.

N. F. Sommer, University of California at Davis, studied postharvest disease pathogens and the effects of radiation on sucrose and fruit juice. A synergistic effect between heat and radiation was noted. When strawberries are treated with hot humid air, the radiation dose can be lowered to 100 krads. The lower dose reduces softening. The induction of abnormal anaphases in *Vicia faba* root-tip cells treated with 2% irradiated sucrose, *p*H > 5, was *not* found. At lower *p*H values, aberrations were noted whether sucrose was irradiated or not.

J. T. R. Nickerson, Massachusetts Institute of Technology (MIT), gave the results of a study of salmonellae in chicken. He exposed salmonellae to gamma rays repeatedly to see if this increased radioresistance. The organisms were exposed 20 times to the radiation; an increase in radioresistance was noted after the 10th cycle. It is not clear whether the increase is due to selection or mutation. An attempt was made to equate virulence with increased resistance to radiation; one of the two strains was more virulent (to a small degree) after 20 cycles of radiation, but the other strain was less virulent. A survey of a chicken processing plant near MIT showed that 64% of the chickens were contaminated, but contamination was on an extremely low level—about 40 organisms per bird.

R. A. Pledger, Hazleton Laboratories, presented the first of many inoculated-pack studies.* A mixture of eight genera of microorganisms is injected into a 10-g sample of chicken. Methods of injection and recovery have been developed; actual measurements are now being made.

J. Liston, University of Washington, tested the effect of split dose vs. single dose on shelf-life extension of English sole fillets and on the factors involved in radiation damage sustained by *Staphylococcus aureus*. A dose of 50 krads to very fresh fish, followed by storage and a second dose of 100 krads, gave the same shelf-life extension as a single dose of 300 krads; the initial dose is best given by a shipboard irradiator. Tracer studies to determine the effect of radiation on RNA and DNA show that the primary lesion is in the DNA molecule. Also, radiation sensitizes cells to further irradiation or heat shock.

J. S. Lee, Oregon State University, undertook to determine what bacteria survive radiation in fish. After a dose of 100 to 300 krads, the surviving bacteria is usually *Achromobacter*. This organism is not well defined taxonomically. His research also showed that total volatile acid and total volatile base produced during shelf life could be used as a measure of spoilage.

A. W. Anderson, Oregon State University, examined *Clostridium botulinum* metabolism for differences between this organism and other *Clostridium* species. Usually, Type E is fermentative and Types A and B are proteolytic. This research has the goal of early and easy detection of *C. botulinum*. Electrophoretic methods for identification of toxin are being developed.

W. P. Segner, Continental Can Co., reported on inoculated-pack studies to assess *C. botulinum* problems in radiation-pasteurized haddock. The study has defined the x value as the time when all members of the panel reject the fish as being spoiled. Segner then presented the protocol of the ad hoc committee concerned with *C. botulinum*. Results at 42°F and lower indicate that there will be no problem with botulism. Odor evaluation alone is apparently not an accurate measure of spoilage, since samples showing mold and slime were acceptable when evaluated only on odor.

J. T. Graikoski, Bureau of Commercial Fisheries, reported toxin production as a result of inoculated-pack studies in yellow perch. Dose was up to 0.2 Mrad; storage temperature was 10°C and below. In nonirradiated fillets and in those irradiated at 0.1 Mrad stored at 10°C, toxin was detected at 12 days; fillets irradiated at 0.2 Mrad were negative for toxin at this time. Again, odor was not a good indicator for spoilage. The length of time toxin persisted after initial production was measured; in some samples, toxin lasted 20 days.

M. W. Eklund, Bureau of Commercial Fisheries, investigated the time of outgrowth of *C. botulinum* in haddock and petrale sole held at 38 to 50°F. Samples were maintained in oxygen-permeable and in oxygen-impermeable wrap. During refrigerated storage the glucose/ribose ratio decreased rapidly in nonirradiated petrale sole fillets but not so rapidly in irradiated samples; *C. botulinum* Type E can utilize either glucose or ribose.

B. Q. Ward, University of Miami, reported that his new contract had, of course, produced no results yet. He will be conducting inoculated-pack studies on shrimp.

C. Ribich, MIT, reported more inoculated-pack studies. Skinless cod fillets were given a dose of 100 to 200 krads, inoculated with 10^2 to 10^6 spores/g, and then stored at 38 to 50°F. No toxin was produced before spoilage at the lower inoculation levels. At higher inoculations, toxin was sometimes produced before spoilage. She pointed out, however, that if a fish were to have 10^4 spores/g in its intestines it would die. The highest contamination found so far has been only 0.17 spore/g. She concluded that radiation pasteurization of fish presents no *C. botulinum* public-health hazard.

N. W. Walls, Georgia Institute of Technology, reported studies of *C. botulinum* Type F. Only three strains have been isolated from natural sources, and these have been used in her studies. The original sample was identified off the coast of Denmark; later Craig found Type F present in salmon from the Columbia River; and Eklund has found Type F in Pacific Ocean finfish. Walls said they have had difficulty producing sporulation, and a correct medium for growth of Type F needs to be determined. Effect of *p*H was noted; using different buffers, they were able to grow Type F at *p*H of from 3 to 10. The high *p*H tolerance might be the result of where Type F is found, i.e., seawater—which has a *p*H of 7.6 to 8.2. *C. botulinum* Type F has also been found recently in Chesapeake Bay crab. Some Type F is proteolytic, but apparently the West Coast sample is not.

D. O. Cliver, University of Wisconsin, has been studying the mutagenesis of four human enteroviruses after irradiation. They were examined for virulence, tissue tropism, host range, and antigens. Tests have been developed to determine these factors. Some differences have been noted, but much more work needs to be done to clarify the situation.

E. F. Reber, University of Massachusetts, gave the first report on wholesomeness studies. Protein quality in clam decreases significantly with increasing radiation dose. In chicken, the effect is the opposite—protein quality increases with increasing

*An inoculated-pack study is a study that is based on the technique of injecting or inoculating sterile samples with specific amounts of bacteria and observing growth under certain conditions.

radiation dose. The presence of thiaminase necessitates the addition of thiamine to any diet using large quantities of freeze-dried clams; even more must be added when raw clams are used in rat feeding studies. He also announced that the extensive bibliography on wholesomeness studies has been published.

H. C. Fegley, AME Associates, reported that the feeding studies using irradiated clams fed to chickens and dogs are nearing completion. No adverse effects have been noted. Some abnormalities were noted in eggs and chicks, but this is felt to be the result of feeding 35% clams rather than an effect of radiation.

K. Morgareidge, Food and Drug Research Laboratories, said that the study using bananas treated with low doses of radiation has just been started. The usual ration of 35% product will be fed to rats and dogs; so far, the 3-month-old pups are eating it well.

P. Derse, Wisconsin Alumni Research Foundation, reported the start of feeding studies using strawberries. No problems have occurred. Some loss of ascorbic acid in the *reduced* form has been noted in the strawberries after irradiation; there has been no loss in the *total* ascorbic acid.

Other Presentations

Dr. Shea summarized the current facilities program of DID, and F. A. Leone reported on DID shipping studies. The chairmen of the four subcommittees then gave reports summarizing research in their fields. L. J. Ronsivalli covered marine products, R. A. Dennison reported on fruit and vegetables, W. P. Segner summarized microbiology, and P. Derse covered wholesomeness tests.

A. T. Spiher, Jr., FDA, gave the status and current thinking at FDA on food-additive petitions. The FDA requires the same kind of data on each and every food additive. Food additives are usually single chemical compounds that have clear-cut effects; they can be added to foods in large amounts in feeding studies and the effects easily noted. The possible presence of radiation-induced chemicals in low concentrations in foods is difficult to detect, and the effects, if any, are subtle. He pointed out that, while data may be insufficient to prove a new additive safe, this does not necessarily indicate that it is unsafe. He again emphasized FDA's rule that raw data are required in the petition to support the scientific conclusions. Much consultation with the Army and the AEC has ironed out most of the former misunderstandings about petition requirements. Spiher noted that the Army has submitted the raw data supporting the ham petition; E. S. Josephson, Natick Laboratories, pointed out that the data filled a 7-ft shelf. S. A. Goldblith asked how long it would take FDA to approve the petition now that they had the data. Spiher would not be specific, but he was encouraging.

After closing remarks by representatives from other government agencies, the contractors' meeting ended.

Study of Irradiated-Pasteurized Fishery Products

By L. J. Ronsivalli and J. H. Carver*

and

Operational Guide for MPDI

By J. D. Kaylor, J. B. Huff, and E. J. Murphy*

Abstract: The annual report to the USAEC by the Bureau of Commercial Fisheries Technological Laboratory, Gloucester, Mass., on radiation pasteurization of marine products is summarized. During the past year, consumer acceptance and optimum dose (250 krads) have tentatively been established for blue-crab meat and scallops. The effect of preirradiation quality on postirradiation shelf life was investigated; cod could be stored 10 days and clam meats 7 days before irradiation became uneconomic. Analytical chemical methods were used to study the odors and flavors in clam meat and haddock. Operation of the Marine Products Development Irradiator was described. A survey determined that product temperatures of less than 40°F were maintained by current fish-distribution methods.

The annual report[1] on the radiation pasteurization of seafoods carried out under a contract from the USAEC's Division of Isotopes Development during the year October 1965 to September 1966 at the Bureau of Commercial Fisheries Technological Laboratory, Gloucester, Mass., is presented here in condensed form. The Marine Products Development Irradiator (MPDI) was used to irradiate the products.

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In research done from 1960 to 1965, the optimum radiation-pasteurization doses and consumer acceptability for air- and vacuum-packed haddock, pollock, ocean perch, and cod fillets and clam meats were determined. Preliminary work showed that mackerel fillets and lobster could also be radiation pasteurized. Studies of the effect of preirradiation quality on postirradiation shelf life showed that even haddock and cod fillets iced for 7 to 10 days prior to irradiation could gain shelf life from the treatment. Many of the available flexible plastic packaging materials with low gas permeability were tested and found suitable for packaging radiation-pasteurized fish.

Evidence from gas-chromatographic and classical wet-chemistry methods showed that radiation enhanced the initial production of volatile carbonyls in fish; however, the final concentration of these compounds in irradiated fish never exceeded the concentration of these compounds found in iced stored nonirradiated products. The development of carbonyls apparently has little to do with product quality in the species tested. Radiation does suppress the development of volatile sulfide compounds that are known to be spoilage indicators, possibly because irradiation at 250 krads kills >99% of the bacteria that are thought to produce the sulfide compounds. Nonetheless, taste-panel data indicate that irradiated fish do eventually spoil and that they become fully as objectionable as spoiled nonirradiated fish.

The research reported was conducted between Oct. 1, 1965, and Sept. 30, 1966, and was performed in two phases. Under Phase I, investigations determined (1) the feasibility of radiation pasteurizing blue-crab meat and scallops, (2) the effect of preirradiation quality on postirradiation shelf life of cod fillets and clam meats, (3) the effects of irradiation and other factors on the organoleptically related chemistry of seafoods, and (4) the microbiological quality of radiation-pasteurized seafoods. A study was initiated to determine the maximum shelf life of both irradiated and nonirradiated seafoods. Under Phase II, work was done at the MPDI on (1) establishing operational procedures, operating limits, and characteristics of the irradiator; (2) conducting and completing a survey of distribution practices in commercial shipments of fresh fish fillets; and (3) cooperating with industry in studies to evaluate the effects on radiation-processed fish fillets shipped in commercial quantities and stored under commercial conditions. A guide was written, covering the most important procedures governing operation and safety at the MPDI.

Radiation Pasteurization of Fishery Products

A radiation dose in the range 150 to 450 krads has been effective in pasteurizing haddock, cod, pollock, ocean perch, soft-shell clam, lobster, mackerel, and blue crab. The species studied so far, the optimum dose for each species, and the shelf-life extensions are shown in Table 1. Other work previously established the acceptability of the irradiated product to the consumer and the effectiveness of several plastic materials for packaging. Radiation at the low pasteurization levels reduces total plate count by 2 to 3 log cycles, and spoilage does not occur until bacteria reach levels of 10^8 per gram. The composition of microflora in irradiated fish is altered significantly.

Table 1 DATA ON RADIATION PASTEURIZATION OF MARINE PRODUCTS FROM THE NORTHWEST ATLANTIC

Storage Temperature = 33°F

Product	Optimum radiation pasteurization dose, krads	Shelf life,* days
Haddock	250	>30
Cod	150	34
Pollock	150	30
Ocean perch	250	30
Mackerel	250	>30
Soft-shell clam†	350	>30
Lobster	150	33

*Average shelf life of nonirradiated samples at 33°F was about 10 to 12 days.
†Vacuum-packed. All others air-packed.

BLUE-CRAB MEAT

The method presently used for preserving much of this product is heat pasteurization. Because the heat-pasteurized product undergoes some color, texture, and flavor losses, radiation pasteurization was investigated as a method offering equal shelf life and better retention of the desirable original organoleptic qualities.

In the first of two experiments, fresh and heat-pasteurized crab meats were obtained from Baltimore 24 to 30 hr after being picked. The fresh meats were irradiated at levels of 150, 250, 350, 450, and 900 krads; they were stored at 33 to 35°F until panel and bacterial evaluation. Results of these tests (Table 2) indicate that blue-crab meat is preserved best by radiation at levels from 150 to 450 krads; the meat irradiated at 900 krads developed definite off-odors and off-flavors. The panel consistently rated the irradiated product higher than the heat-pasteurized product with respect to appearance, odor, and flavor.

In the second experiment, some fresh crab meat was heat-pasteurized by Bureau of Commercial Fisheries (BCF) workers; in all other respects the two experiments were identical. Crab meat irradiated at 150 krads spoiled by the 103rd day of storage, but the other samples remained unspoiled until they were all used up on the 180th day. The optimum dose recommended for preserving blue-crab meat is 250

Table 2 RESULTS OF OPTIMUM-DOSE-LEVEL STUDIES ON PICKED BLUE-CRAB MEAT AND STANDARD TOTAL AEROBIC PLATE COUNTS (TPC) ON THESE MEATS AT THE END OF STORAGE PERIOD

Treatment	Experiment 1		Experiment 2	
	Storage, days	TPC/g	Storage, days	TPC/g
Nonpasteurized	10–13*	600×10^6		
150 krads	>56†	$<3 \times 10^5$	85 to 102	56×10^6
250 krads	>56†	2.8×10^6	>180†	49×10^3
350 krads	>56†	3×10^3	>159†	1500
450 krads	>56†	<300	>180†	<300
900 krads	21–29			
Commercially heat-pasteurized	>56†	<300	>180†	<300
Laboratory heat-pasteurized			>180†	23×10^4

*Two-day-old unpasteurized meats were coliform positive. Presumptive tests for MPN (most probable number) coliforms of heat-pasteurized and irradiated meats, taken during and at the end of storage, were consistently negative.
†Samples were depleted on this day.

krads. Bacteriological examinations indicated that when samples did spoil the number of aerobic organisms was greater than 10^6 organisms/g; samples irradiated at doses of 350 krads and above had very low counts throughout the test period. However, it was apparent that the microorganisms surviving the 250-krad level of irradiation had little effect on the organoleptic quality of the product.

SCALLOPS

Fresh scallop meats were frozen for reference samples or packed and irradiated at doses of 250, 500, 1000, 2000, and 4500 krads. Irradiated samples were stored at 33 to 35° F. After 2 weeks, samples were evaluated raw, steamed, and fried (Table 3). In general, the irradiated samples were less acceptable than the nonirradiated samples; this was especially true of scallops irradiated at doses above 1000 krads. Deep-fat frying tended to hide off-odors and off-flavors induced by high radiation doses.

A second experiment was undertaken to examine the effect of packing in tins under vacuum. Again, some scallop meat was frozen for use as reference samples. Half the remainder was canned in air-pack and half was canned under vacuum. Radiation doses were 250, 500, 750, and 1000 krads. Taste-panel results showed that irradiated scallops were acceptable after 55 days' storage; nonirradiated scallops spoiled between the 17th and 20th days of storage. No organoleptic differences were noted between air-packed and vacuum-packed samples. Storage time was almost three times that of nonirradiated samples, and, since all dose levels were effective, an optimum dose of 250 krads (the lowest level used) is tentatively recommended for sea scallops.

Table 3 ORGANOLEPTIC RESULTS ON RAW, STEAM-COOKED, AND DEEP-FAT-FRIED SCALLOPS STORED UP TO 10 DAYS AT 33 TO 35°F

Cooking method	Postirradiation test day	Scores* of nonirradiated frozen scallops	Scores of irradiated scallops				
			Dose, krads				
			250	500	1000	2000	4500
Raw	1	4.5	4.1	4.0	3.8	3.5	3.2
	2	4.4	3.9	3.5	3.6	3.3	3.1
	3	4.4	3.8	3.4	2.8	3.3	2.5
	4	4.5	3.7	3.6	3.6	3.4	2.8
	5	4.4	3.8	3.3	3.4	2.9	2.7
	6	4.5	3.7	3.4	3.5	3.5	3.0
	7	4.2	3.5	3.7	3.4	3.3	3.3
	8	4.5	3.6	3.4	3.3	3.7	2.8
	9	4.8	3.7	3.4	3.3	3.3	2.8
	10	4.8	3.7	3.3	3.5	3.3	2.8
Steamed	1	4.2	4.1	3.5	3.5	2.7	2.1
	2	3.8	3.6	3.5	2.3	2.3	1.6
	3	3.8	3.9	3.4	2.7	2.3	1.5
	4	3.9	3.4	3.4	2.5	1.8	1.5
	5	3.9	3.1	3.1	2.7	2.5	1.4
	6	4.0	3.4	3.1	2.9	1.9	
	7	3.7	3.0	3.6	2.4	2.0	
	8	3.8	3.1	3.2	2.6	1.7	
Deep-fat-fried	1	4.2	3.9	3.8	3.8	3.4	2.9
	2	4.2	3.9	3.2	3.8	4.1	2.6
	3	3.9	4.0	4.2	3.6	3.4	2.8
	4	3.9	3.3	3.6	3.7	3.4	3.0
	5	3.2	3.9	3.8	3.4	3.9	2.5
	6	3.8	4.2	3.9	3.6	3.3	2.4
	7	4.0	3.8	3.9	3.1	2.6	2.9
	8	4.0	3.8	3.7	3.7	3.0	2.3
	9	3.9	3.9	3.5	3.5	3.6	2.2
	10	4.0	3.6	3.4	3.3	2.6	2.5

*The 5-point scale was used where 5 = excellent, 4 = very good, 3 = good, 2 = fair, and 1 = poor.

ACCEPTANCE STUDIES

Experiments were conducted at several locations to determine whether or not frozen clam samples could be used as reference samples in the study of radiation pasteurization of clams and to determine whether or not U. S. Army personnel would eat irradiated or nonirradiated clams.

In previous BCF work on radiation pasteurization of finfish, fresh frozen finfish were used as reference samples. Fresh frozen clam meat was therefore evaluated for use as reference samples in radiation-pasteurization studies of clam meat. Half of a lot of freshly shucked clams was frozen; the other half was given a dose of 450 krads and stored at 33° F. After various periods of storage, portions of each half were evaluated raw, breaded, and deep-fat fried. Taste-panel results showed that the frozen clam was more acceptable than the irradiated sample and only slightly less acceptable than a nonfrozen sample. All three kinds of samples were acceptable. On this basis, frozen clam meats appear to be satisfactory as experimental reference samples.

Pioneer Research Laboratory. Radiation-pasteurized clam meats were tested by taste panels at the Pioneer Research Laboratory, U. S. Army Natick Laboratories, Natick, Mass. Results of evaluation by consumer panels of 36 to 40 members and technical panels of 8 to 10 members indicate that clams stored 3 and 4 weeks after irradiation are as acceptable as clams that have been stored frozen.

Fort Lee, Va. A large-scale consumer acceptance test was conducted by serving irradiated and frozen soft-shell clam meats to U. S. Army personnel at Fort Lee, Va. Nonirradiated fresh haddock fillets were also served to the troops to provide a reference material since most of the soldiers were presumed to be unfamiliar with clam meat, which is chiefly a New England seafood. Results[2] of these tests (Table 4) indicate that irradiated and nonirradiated clam meats were equally acceptable after 3 weeks' storage; after 4 weeks' storage, irradiated clam meats were less acceptable. All clam meats were less acceptable than haddock fillets throughout the test.

Effect of Preirradiation Quality

The effect of preirradiation quality on postirradiation storage of cod fillets and soft-shell clam meats was studied. Since radiation pasteurization—like any other preservative process—does not improve the quality of the product, application of the process would not be economic if at the time of irradiation the quality of the product were marginal. A minimum extension of 7 days in shelf life is considered necessary for radiation processing of marine products to be economic.

COD

Half of a large lot of freshly caught codfish was eviscerated, filleted, packaged, and frozen; the other half was well-iced and stored at 42° F. On the 2nd, 4th, 6th, 8th, 10th, 12th, and 15th days of storage, some of the iced fish were filleted, packaged, irradiated, and stored at 33 to 35° F. At periodic intervals the irradiated cod fillets were evaluated raw and steamed. In one experiment, irradiated fillets were further evaluated after deep-fat frying to determine the masking effect of this cooking method. Results (Table 5) indicate that irradiated fillets prepared from fish held on ice for 10 days prior to irradiation had a storage-life extension of 10 days. No appreciable storage-life extension was realized by radiation pasteurization when cod was stored on ice for 12 days or more before irradiation. Previously, an experiment[3] showed that iced haddock could be irradiated after 9 days' storage and gain a significant shelf-life extension. The data indicate that,

Table 4 AVERAGE HEDONIC RATINGS FOR IRRADIATED AND NONIRRADIATED CLAMS, NONIRRADIATED HADDOCK, AND PERCENT OF EACH ITEM CONSUMED*

	Clams						Haddock		
	Irradiated			Nonirradiated			Nonirradiated		
Company	Men rating, No.	Average rating†	Amount eaten, %	Men rating, No.	Average rating	Amount eaten, %	Men rating, No.	Average rating	Amount eaten, %
135th QM Co.	43	5.53	74.2	84	5.44	80.5	132	6.69	60.7
Spec Troops	103	4.54		106	4.94		215	5.62	
596th QM Co.	98	4.77	62.7	59	4.68	80.1	160	5.84	95.1
1st week summary	244	4.81	66.5	249	5.05	80.3	507	5.97	82.1
WAADS	67	4.37	74.6	59	5.17	84.2	139	6.45	91.4
Hq. QMSTC	96	4.14	57.8	90	5.78	61.4	176	5.69	78.8
22nd FASCOM	59	5.15	73.3	193	4.63	66.7	214	5.57	82.5
2nd week summary	222	4.48	67.6	342	5.03	70.8	529	5.84	84.0
Overall summary	466	4.65	67.2	591	5.04	74.1	1036	5.90	83.5

*This table taken from Ref. 3.

†The 9-point scale was used where 9 = like extremely, 8 = like very much, 7 = like moderately, 6 = like slightly, 5 = neither like nor dislike, 4 = dislike slightly, 3 = dislike moderately, 2 = dislike very much, and 1 = dislike extremely.

in general, the fresher the fish prior to irradiation the longer the total refrigerator storage life. Also, the data show the masking effect of deep-fat frying. Cod fillets judged marginal when raw and unacceptable when steamed were acceptable 39 days later when deep-fat fried.

Table 5 POSTIRRADIATION SHELF LIFE OF FILLETS CUT FROM COD OF VARYING QUALITY, IRRADIATED AT 150 KRADS, AND STORED AT 33 TO 35°F

Preirradiation storage time on ice, days	Postirradiation storage time, days		Total storage time, days	
	Steamed cod*	Deep-fat-fried cod†	Steamed cod*	Deep-fat-fried cod†
2	25	62	27	64
4	18	65	22	69
6	15	60	21	66
8	15	40	23	48
10	10	34	20	44
12	0	39	<12	51
15 (spoiled)	0	0	<15	<15

*Average of data from two experiments.
†Data from one experiment.

CLAM MEATS

Freshly shucked clam meats were stored in a refrigerated room kept at 33 to 35° F. On the 1st, 3rd, 5th, 7th, 9th, and 11th days of storage, some clam meats were canned, irradiated, and stored further at 33 to 35° F. Periodically, a taste panel evaluated the clam meats raw and after deep-fat frying. Results showed that clam meats are suitable for radiation pasteurization when stored on ice up to 7 days prior to irradiation. There was some texture loss, and clam meats adjacent to headspace air in the cans darkened. The darkening, an oxidative reaction, can be prevented by vacuum-packing or by filling the can completely; the texture change is probably due to autolysis.

Microbiological studies indicate that the 450-krad dose used destroyed nearly all aerobes. The product eventually spoiled as a result of one or a combination of (1) anaerobic bacteria, (2) endogenous enzymes, (3) enzymes produced by aerobic bacteria prior to irradiation, or (4) other oxidative degradation.

Flavor and Odor Studies

Applied chemistry studies have been conducted to attempt to identify the chemical entities responsible for the flavor and odor of fishery products and to follow the changes in concentration of these compounds occurring as a result of radiation pasteurization, storage time, and storage temperature.* Seafoods whose chemistry was studied during the period covered by this report include soft-shell clam meats and haddock and cod fillets, both irradiated and non-irradiated. Most of the work was concerned with obtaining gas-chromatographic scans of the odor compounds from these seafoods and determining changes resulting from various treatments; a new study was started to determine differences in spoilage patterns resulting from endogenous chemical reactions and from bacterial action.

CLAM MEATS

More than 20 volatile compounds (Table 6) from clam meats previously differentiated by gas chromatography were positively identified by time-of-flight mass spectrometry.

Table 6 COMPOUNDS IN TOTAL VOLATILE CONDENSATES OF CLAM MEATS POSITIVELY IDENTIFIED BY TIME-OF-FLIGHT MASS SPECTROMETRY

1. Hydrogen sulfide	12. Isovaleraldehyde
2. Carbon disulfide	13. Benzene
3. Dimethyl sulfide	14. Toluene
4. Dimethyl disulfide	15. Methyl ethyl ketone
5. Dimethyl trisulfide	16. Methyl acetate
6. Methyl mercaptan	17. *n*-Octane
7. Ethanol	18. *n*-Heptane
8. Butanol	19. Butene
9. 3-Methyl 1-butanol	20. 1-Pentene
10. 2-Methyl furan	21. Hexene
11. Acetone	22. Trimethylamine

A second study on clam meats was conducted to determine the effects of irradiation, cooking, storage, packaging, and combinations of these on the volatile carbonyl compounds in this product. Fresh clam meats were packed in cans; half were air-packed and half were vacuum-packed. After irradiation (450 krads for air-packed and 350 krads for vacuum-packed), clams were evaluated after 2, 9, 20, and 30 days of storage at 33 to 35° F. Some were cooked, and some were left raw. The concentration of nearly every detectable compound from air-packed samples stored 2 days was increased by irradiation. Cooking

*Many of the analytical procedures and techniques used for this work were developed under a separate contract with the USAEC's Division of Biology and Medicine.

had a similar overall effect except that it reduced the amount of low-molecular-weight carbonyls. Volatile carbonyls in nonirradiated samples, cooked and raw, increased during storage, reaching a maximum sometime between the 9th and 30th days of storage. The rate of carbonyl formation in irradiated, raw, air-packed samples was accelerated, with the concentration reaching a maximum between the 2nd and 9th days of storage. The immediate combined effect of irradiation, storage, and heating on air-packed clams is evidently a substantial initial increase in carbonyl concentration. The carbonyl concentration in the samples stored 20 and 30 days was markedly decreased, however. Apparently, the whole development–dissipation cycle of carbonyls is accelerated. The same general patterns of carbonyl development were noted in the vacuum-packed samples as in the air-packed samples; however, because free oxygen plays a role in the development of volatile carbonyls, carbonyl concentrations were much lower in the vacuum-packed samples.

Although irradiation delays the onset of spoilage, it accelerates the development of carbonyls. This apparently means that carbonyls have little connection with spoilage.

HADDOCK

Work has started on correlating results of panel evaluations of odors from irradiated and nonirradiated haddock fillets with gas chromatographic scans on total volatile compounds. Generally, irradiation caused new volatile components to be formed and an increase in the number of detectable volatile components. In some cases, volatiles present in nonirradiated samples were not detected in irradiated samples. Full evaluation of scans at this time is difficult, and no conclusions will be made until after volatiles are positively identified on a time-of-flight mass spectrometer.

Technique for Excising Sterile Fish

Changes in volatiles are a result of bacterial action and of autolysis. To determine those changes caused only by autolysis requires the analysis of sterile fish. A technique for excising and aseptically handling sterile flesh from fish has been developed; sterile samples are recovered in 25% of the attempts, but experience is expected to improve the recovery percentage.

Chromatograph scans of volatiles showed about 20 compounds from fresh cod, about 30 compounds from nonsterile cod stored 21 days, about 25 compounds from sterile cod stored 21 days, and about 42 compounds from nonsterile cod stored 30 days. Positive identification of these compounds will be necessary before any conclusions about the compounds can be reached.

Microbiology

Bacteriological services for technologists in other projects of the radiation preservation program were provided; total plate counts conducted concurrently with organoleptic analyses support earlier data that indicated radiation reduced the total plate counts by about 2 to 3 log cycles.

Maximum Storage Life of Haddock Fillets

Two experiments were set up to determine the absolute maximum shelf life of radiation-pasteurized and nonirradiated haddock fillets; rejection was determined by odor of the raw fillets. Samples received doses of 0, 50, 100, or 200 krads. The samples in the first experiment were stored at 33, 40, 46, 50, and 60° F; the samples in the second experiment were stored at 33, 40, 46, 55, and 60° F. The maximum shelf life of a sample was considered reached when an untrained panel of about 20 members rejected the sample. Samples were also evaluated by a trained panel, who usually rejected samples earlier than the untrained panel because they could identify objectionable odors earlier. The results are shown in Table 7. Evidently a dose of 50 krads has little preservative effect. Samples irradiated at 100 and 200 krads attained relatively high plate counts before being rejected; in two cases the count reached 10^{10} and 10^{11} organisms/g. The conclusions reached so far: (1) shelf life increases with increasing dose and decreasing storage temperature; (2) at storage temperatures above 40° F, maximum shelf life is not increased by packaging irradiated fillets in impermeable plastic films; (3) there appears to be no significant difference in degree of offensiveness between spoiled irradiated fillets and spoiled nonirradiated fillets; (4) total aerobic plate counts of rejected irradiated fillets are relatively higher than counts of rejected nonirradiated fillets; and (5) the relatively long maximum shelf lives obtained were the result of the strict definition of shelf life used in the study, i.e., time until rejection by all members of an untrained panel.

Market Life of Cod Fillets

The end of the market life of a product is that time at which product quality has degraded until consumer acceptance is borderline. To develop information on market shelf life of irradiated products held at other than ideal storage temperatures, very fresh cod were irradiated at 150 krads and stored at 33, 37, 42, and 47° F. Results (Table 8) appear to substantiate the finding that irradiation effectiveness is increased as the storage temperature is lowered. Actually all fishery products should be stored at temperatures as near to freezing as possible.

Table 7 MAXIMUM SHELF LIFE, DAYS, OF IRRADIATED AND NONIRRADIATED RAW HADDOCK FILLETS BASED ON ODOR EVALUATION

Temp., °F	Packaging material*	Maximum shelf life, days, of haddock fillets											
		0 krads			50 krads			100 krads			200 krads		
		T†	L‡	C§	T	L	C	T	L	C	T	L	C
60	P	2	4	5				5	6	7	7	10	10
		2	3	4				6	6	7	6	7	8
	PP	2	4	5				5	7	7	7	10	10
		2	4	4				5	6	7	7	7	11
55	P	2	4	5				7	10	10	7	10	11
	PP	2	4	5				7	10	10	9	11	12
50	P	3	7	7				6	12	13	11	18	22
	PP	3	7	7				8	12	13	12	13	13
46	P	6	7	11				9	11	13	12	18	25
		11	13	13	7	8	11	8	12	14	8	14	15
	PP	5	7	12				10	11	12	12	20	21
		8	8	11	11	11	11	7	8	11	19	27	27
40	P	10	11	14	13	13	17	24	39	39	24	27	27
		8	14	15	10	22	22	20	20	20	21	22	24
	PP	10	11	18	13	13	14	24	39	39	35	49	55
		8	15	16	20	21	21	20	28	28	45	49	59
33	P	20	39	39	26	49		41	52		56	62	
		24	31	34	38	38		38	49		42	42	
	PP	20	45	46	33	49		49	52		56	62	
		23	29	29	38	42		38	45		52	59	

*P designates polyethylene; PP designates polyethylene-coated polyester.
†T is the rejection time, days, by the trained panel (12 members).
‡L is the rejection time, days, by the untrained panel in the laboratory (20 to 30 members).
§C is the rejection time, days, by the untrained panel at the Massachusetts Institute of Technology (about 20 members).

Two chemical tests were conducted on both irradiated and nonirradiated fish; these were the thiobarbituric acid test (TBA) for rancidity and the total volatile acid test (TVA) for degradative products. There were no changes in the TVA and TBA values in irradiated fish that could be correlated with changes in organoleptic quality.

Table 8 MARKET STORAGE LIFE OF COD FILLETS*

Storage temp., °F	Bulk-packaged, nonirradiated, days	Bulk-packaged, irradiated, days	Air-packed, canned, irradiated, days
33	10	28	31
37	6	10	11
42	4	9	6
47	2	5	4

*Adapted from Fig. 19 of Ref. 1.

Operation of the Marine Products Development Irradiator

In operation since April 1965, the Marine Products Development Irradiator (MPDI) installation[4] is a one-story building containing a refrigerated storage room, a product conveyor, a gamma-radiation cell, and auxiliary areas consisting of office, health physics, dosimetry laboratory, and general work and storage space. Industry's participation in irradiation studies is encouraged; irradiation requests from university, federal, and state research groups are also handled. A considerable amount of information on the shipping and marketing of fresh fishery products has been compiled to determine the actual operating conditions of handling fishery products; the temperatures and quality retention of both irradiated and nonirradiated fillets under actual operating conditions have been evaluated. The overall supervision of the MPDI is the responsibility of a scientist, licensed by the USAEC, who is assisted by a licensed health physicist, a physical science technician, and a physical science aide.

Since the MPDI is a prototype, modifications to improve its performance are inevitable. The radiation-source elevator was designed to operate with stainless-steel bearings in stainless-steel races without lubrication, because radiation was thought to cause breakdown of lubricants. When replacement was necessary, conventional lubricated roller bearings[5] were installed to facilitate operation. The fast and slow conveyors that carry product into and out of the gamma cell have been refined to provide larger tolerances than were originally designed into the system.

PROCESSING

Commercially prepared fillets are packed in 10-, 20-, or 30-lb capacity fillet tins, either in Food and Drug Administration (FDA)-approved film wrap or in bulk, and then refrigerated. Before processing, each fillet tin is tagged with an indicator that changes color

from yellow to red when irradiated. The tins are fed into the gamma cell by the fast conveyor, which transfers them to a slower conveyor for the actual irradiation. To ensure dose uniformity, each tin makes two complete round trips through the gamma cell on each of both sides of the source center line.

RADIATION SOURCE

The original source of 235 thousand curies of ^{60}Co has decayed to 160 thousand curies (Fall 1966). The source is made of 6 replaceable units, each containing 16 Brookhaven National Laboratory Mark I ^{60}Co strips. Rated source utilization is about 21%, using target overlap for maximum efficiency. The normal dose for fillets is about 200 krads at a production rate of 1 ton/hr with a maximum/minimum dose ratio of 1.3.

DOSIMETRY

Absorbed dose is measured by using a Masonite duplicate of the fillet tin; the phantom has 27 rabbets for positioning dosimeters uniformly. The dosimeter used is the chemical ferrous sulfate–cupric sulfate system, accurate to ±1% from 40 krads to 700 krads, and it gives results[6] with a reproducibility of ±0.3%. The solution used is a 0.005*M* H_2SO_4 aqueous solution, 0.001*M* in ferrous sulfate and 0.01*M* in cupric sulfate. The amount of radiation absorbed is determined by measuring the change in optical density of the solution at a wavelength of 304 mμ at 25°C.

MEASUREMENT OF DOSE

Title 21, of *Food and Drugs*, Chap. I, Food and Drug Administration, Department of Health, Education, and Welfare, Subchap. B, Food and Drug Products, Paragraph 121, Food Additives, Subparagraph G states:

> A record of total dose absorbed shall be obtained by the use of phantoms having the same geometry as the containers of food and containing dosimeters suitable for the maintenance of a permanent record of exposure. Measurements of total dose shall be made by the use of one phantom per 24 hour period of exposure or by use of one phantom for each thousand packages exposed whichever occurs first, with the exposure records being retained for Food and Drug Administration inspection for a period of one year.

This rule is followed as a matter of course in MPDI operations.

Minimum dose is measured from dosimeters placed in the central layer of the phantom; maximum dose is obtained from dosimeters on the surface of the phantom. Loaded phantoms are irradiated along with products, and only solution less than 8 hr old is used in the dosimeters. Records are maintained for 1 year.

RADIATION SERVICES

Although the MPDI was designed primarily for irradiation of seafoods, it is often used for other radiation services. This past year, radiation services were provided for several state and federal agencies and industry.

In cooperation with the Plant Pest Control Division of the U. S. Department of Agriculture (USDA), several thousand male pupae of the gypsy moth were irradiated and then mated with normal females. Some were marked with a special dye and released with chemically sterilized males to compete in a forest; the object was to determine if the larger laboratory-bred pupae would be more sexually aggressive than the smaller, normal males. The study indicated that 20 krads reduced hatchability of eggs by over 99.6%; doses greater than 27.5 krads resulted in mortality or physical defects. Results of three separate jobs will be published by USDA.

For use in proposed 90-day animal-feeding studies, 3500 lb of Hawaiian papayas were irradiated. Low-level irradiation of papayas effectively controls a fruit fly that is a potential pest if introduced into the mainland United States. The current control method is a special hot-water and fumigation treatment that lasts 30 hr.

Hawaiian mangoes may not be imported into the mainland United States because of a weevil that grows inside the seed of mangoes and cannot be killed by fumigation or other external treatment. Gamma rays at low levels are extremely effective in killing the insect without affecting the fruit. The MPDI irradiated 4000 lb of Florida mangoes, in place of Hawaiian mangoes, for use in animal-feeding studies.

In cooperation with the Commonwealth of Massachusetts Department of Natural Resources, the possibility of irradiating clams taken from grossly polluted waters was investigated. Results indicate that 50 krads will significantly reduce the coliform count; a median reduction of over 97% was obtained. The level of radiation did not affect the living clam throughout the holding period, which was greater than that normally encountered in commercial outlets.

The amount of material irradiated during the period of this report is quite small when compared to the capacity of the MPDI. There is little advantage to be gained from irradiating more material than can serve a useful purpose, since irradiated fishery products have not yet been cleared for general sale. However, during the report period, the MPDI has completed 120 irradiation jobs for a total of 43,300 lb of products (Table 9). These were mostly seafoods but also included fruits, meats, chicken, nonfood products, and three insect sterilization studies.

Distribution Survey

The fresh-fish fillet industry of 1966 remained in virtually the same degree of simplicity as it did in 1926. Because of the lack of published data on the effectiveness of the distribution system in maintaining low product temperatures, a survey was undertaken

Table 9 IRRADIATION SERVICES PERFORMED AT THE MARINE PRODUCTS DEVELOPMENT IRRADIATOR—OCT. 1, 1965 TO SEPT. 30, 1966

Product	No. of jobs	Quantity	Purpose
Clams, live	1	33 bags	Research, pasteurization
Clams, sea	1	25 lb	Research, pasteurization
Clams, shucked	6	28,160 lb	Research, pasteurization
Chicken	1	80 lb	Research, pasteurization
Cod, fillets	16	948 lb	Research, pasteurization
Cod, whole	1	25 lb	Research, pasteurization
Crab meat, blue	1	40 lb	Industry, pasteurization
Crab meat, Dungeness	1	20 lb	Research, pasteurization
Crab meat, king	1	15 lb	Research, pasteurization
Culture medium	1		Research, sterilization
Film, bags	4	600 lb	Research, sterilization
Fish extract	1	7 tubes	Research, pasteurization
Fish oil	3	10 lb	Research, sterilization
Flounder	2	60 lb	Research, pasteurization
Flounder	2	540 lb	Industry, pasteurization
Glassware	1	Assorted	Research, pasteurization
Gypsy-moth pupae	3	3,571 pupae	Sexual sterilization
Haddock, fillets	30	3,175 lb	Research, pasteurization
Haddock, fillets	11	1,885 lb	Industry, pasteurization
Ham	2	360 lb	Research, sterilization
Mangoes	1	4,000 lb	Research, pasteurization
Microorganism	1	10 cultures	Research, pasteurization
Oyster, live	1	28 lb	Research, pasteurization
Oyster, shucked	3	27 lb	Research, pasteurization
Papayas	1	3,500 lb	Research, pasteurization
Perch, ocean	2	70 lb	Research, pasteurization
Perch, yellow	1	10 lb	Industry, pasteurization
Perch, yellow	1	25 lb	Research, pasteurization
Pike	1	10 lb	Industry, pasteurization
Scallops	3	260 lb	Research, pasteurization
Scallops	2	50 lb	Research, pasteurization
Scallops	1	5 lb	Industry evaluation
Seawater	4	4 bottles	Research, pasteurization
Wood–plastic compound	2	Various	Industry, polymerization

to determine temperature patterns of fresh fillets shipped in commercial quantities under commercial conditions. Unless current industry practice maintained product temperatures of less than 40° F, the prospects for commercialization of fish irradiation would not be promising.

The survey began in January 1965. At first, industry allowed their regular commercial shipments to be measured; later, fillets especially prepared for the study were shipped under commercial conditions. A temperature-sensing probe was inserted into the center of the haddock fillet containers, and the leads were run to the outside of the bulk shipping container. Vehicle air temperatures and outside air temperatures were also obtained. An observer accompanied each shipment to record temperatures and make observations on handling practices. When the destination was reached, the observer shipped all samples back to Gloucester by airfreight for laboratory testing.

Fresh-fish shipments are made in boxes to outlets located no further away from Boston than Florida and Ohio and in barrels to outlets located at greater distances. Boxes are transported by refrigerated trucks, and barrels are iced and transported by unrefrigerated boxcars. Two shipments were made during each season of the year to embrace all extremes of weather conditions.

FROZEN-FOOD TRUCKS

Well-insulated, mechanically refrigerated trucks make long-distance hauls of both frozen and iced fish to Kentucky. Trips take 5 or 6 days, depending on the number of delivery stops scheduled. Frozen fish is kept in the main section of the trailer with boxed iced fish stored in a small section at the rear, separated from the frozen fish by a plastic or canvas curtain or sometimes a 4-in. insulated wall. Usually the iced fish become partially frozen before the end of the trip. Fresh-fish temperatures invariably go below 32° F on these trips.

PROCESSOR–DISTRIBUTOR TRUCKS

Some fish processors act as their own distributors. The one investigated here has a small fleet of trucks and makes sales in western Massachusetts and eastern New York. Weekly round trips take 24 to 30 hr. The trucks are compartmented and well insulated. Iced fresh seafoods are transported in the forward compartment; frozen seafoods are trans-

ported in the rear compartment with temperature maintained by electrically precooled plates. The temperature of fillets is higher than desirable at the beginning of the trip; the temperature drops during the distribution trip but not sufficiently because of the frequent opening of the compartment doors.

REFRIGERATED FRESH-FISH TRUCKS

Fresh seafood is shipped from Boston to Ohio several times a week in well-insulated, mechanically refrigerated trucks. Experience has taught the truck drivers to set their mechanical refrigeration to lower the trailer temperature to 28° F. The combination of ice surrounding the fillet tins plus the refrigeration temperature serves to keep the product right at the freezing point of water after 2.5 days of travel. When the product arrives at its destination, product temperature is ideal; this is true regardless of product temperature at the start of the trip.

RAILWAY EXPRESS

Transporting by rail—perhaps the oldest commercial fish distribution method—is accomplished by icing five 20-lb fillet tins in 150 lb of ice in a clean wooden barrel. A combination burlap–plastic flexible cover is coopered over the barrel top. The product is carried in noninsulated, nonrefrigerated general freight cars; temperatures frequently rise into the 80's in these cars. Barrels are reiced in transit if the destination is sufficiently distant. This primitive method is quite efficient.

Commercial Benefit Studies

In April 1966, with FDA approval, seafood buyers of national, regional, and local chain stores were approached and acquainted with the principles of radiation pasteurization of seafood. They agreed to evaluate samples of irradiated and nonirradiated fillets and to comment on the results. No sales of either irradiated or nonirradiated fillets were made in the experiment. Only qualified testers participated, and all samples were destroyed at the end of the experiment. After they had determined for themselves that the irradiated samples outlasted the control samples from a little less than a week to as much as 2 weeks, the seafood buyers wanted to know how soon clearance could be obtained despite the fact that they were told specifically that clearance from FDA would not be given for at least 2 years.

The buyers are convinced that the process has commercial application, although most of them saw no particular advantage in having a shelf-life extension of much more than a week over present nonirradiated fish. Retailers claim that use of radiation-pasteurized fish would greatly reduce the present markdown and unavoidable loss of fresh seafoods after the Friday peak. Large chain supermarkets state they could and would sell irradiated fresh seafoods where they now sell no fresh seafoods; new markets appeal to them.

Some industry members expressed dismay over having to label the product as irradiated, but others thought that government approval would allay consumer apprehension. The amount of concern about consumer reaction was in direct proportion to level of management; the higher the management, the greater the concern.

Generous and unsolicited offers were received from supermarkets offering their facilities for any retail testing, but since FDA clearance had not been obtained, the offers were not accepted. This is, of course, the usual and practical way of determining consumer attitudes, reactions, and volume of repeat sales. The consensus of chain-store seafood buyers is that irradiation of seafoods will: (1) cause seafood to remain fresh longer, (2) reduce waste, (3) enable stores to pass savings along to consumers, (4) allow for not less than usual profits, and (5) open up new market areas.

Description of Appendices

The Bureau of Commercial Fisheries annual report has appendices detailing methods for taste-panel evaluations and for analysis of volatiles in fish; a proposal for a study on *Clostridium botulinum* Type E hazards in irradiated fish is also included. Finally, Appendix IV is the Marine Products Development Irradiator Operational Guide. It lists administrative procedures and routine operation procedures step by step; it also details nonroutine operations procedure and describes the radiation protection program used at the MPDI. (FEM)

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EFFECT OF IRRADIATION ON THE MICROBIAL FLORA SURVIVING IRRADIATION PASTEURIZATION OF SEAFOODS, ANNUAL REPORT, APRIL 1, 1966–MARCH 31, 1967, by R. O. Sinnhuber and J. S. Lee, USAEC Report TID-24171, Apr. 15, 1967 (CFSTI, 67 pp.)

The annual report to the Division of Isotopes Development, USAEC, for the period Apr. 1, 1966, to Mar. 31, 1967, gives results of research performed on the survival primarily of *Achromobacter* in irradiated seafoods. The report is presented in three parts. Part I provides a characterization of the genus *Achromobacter*, a heretofore poorly defined group. Part II gives results of an investigation of the chemistry of spoilage of irradiated and nonirradiated seafoods based on inoculated pack studies. In Part III, the results of a comparative study of preservation methods are given; gamma radiation was compared to the application of chlortetracycline (CTC) and to freezing for effect on the microbial population of ocean perch.

The major findings during the period reported were as follows: (1) The optimum growth temperature for *Achromobacter* species was between 24 and 28°C. However, some species were able to grow at all temperatures between 0 and 37°C. (2) All *Achromobacter* species tested could tolerate NaCl concentrations up to 7.5%. (3) Radiosensitivity of *Achromobacter* species varied by about a factor of 10, ranging from 5.0 to 75.0 krads for an LD_{50} in 0.067*M* phosphate buffer at a *p*H of 7.0. *Achromobacter* species isolated from irradiated seafoods did not have a higher radioresistance than those isolated from nonirradiated seafoods. (4) The total volatile bases and total volatile acids produced in fish inoculated with *Pseudomonas* (PI-950) and *Achromobacter* did not differ markedly. However, production of total volatile bases in fish that spoiled naturally was higher than in fish inoculated with PI-950, and inoculation with PI-950 did result in higher production of total volatile acids. In all experiments a microbial count of 10^8/g had to be reached before an increase in production of total volatile bases was noted. (5) Microbial flora of ocean perch changed very little when treated with CTC or freezing, but CTC treatment resulted in buildup of CTC-resistant microbial groups. (6) *Achromobacter* species were the dominant survivors in ocean perch that had been given a dose of 0.1 Mrad. (FEM)

USE OF RADIOISOTOPES IN CHEMISTRY

The literature through 1965 (523 references, about half Russian and half Western European and American) on the use of radioisotopes in analytical, inorganic, physical, and organic chemistry has been reviewed in a Russian book.* The principles of radioisotope use and methods for making radioisotopes and labeled compounds are discussed by way of introduction. Subjects covered include the use of radioisotopes as tracers for studying both physical and chemical reactions, catalysis, surface area and particle dimensions in precipitates, solubility, and saturated vapor pressures; in radiometric titrations; and in activation analysis and isotope dilution work. Radioisotopic instruments based on both absorption and backscattering of radiation for quantitative determination of the components of a mixture are described. (MG)

*A. N. Murin (Ed.), *Radioactive Isotopes in Chemical Research* (in Russian), Izdatel'stvo "Khimiya," Leningrad–Moscow, 1965 (300 pp.).

RECOVERY OF ACTINIDE ELEMENTS

An extensive review* of the literature (420 references) on recovery of actinide elements was made by Ulstrup of the University of Denmark in Lyngby and published by the International Atomic Energy Agency. The elements discussed are actinium, thorium, protactinium, uranium, neptunium, plutonium, americium, curium, nobelium, and lawrencium. Both the conventional separation processes usually used in preparation of these nuclides—precipitation, organic solvent extraction (including chelation and complexing), and ion exchange—and the less common methods used in analytical work and of possible interest for preparation, e.g., electrophoresis and electrolysis, chromatography, amalgamation, pyrometallurgy, and volatilization, are discussed, some in detail. A less extensive review (70 references) of methods for making targets to be irradiated, irradiation conditions, and nuclide recovery was published in France† a few months earlier. (A brief summary‡ of Oak Ridge National Laboratory work in this field was printed in a recent issue of *Isotopes and Radiation Technology*.) (MG)

*J. Ulstrup, Methods of Separating the Actinide Elements, *Atomic Energy Review*, 4(4): 3-82 (1966).

†R. Berger, Possibilities of Manufacturing Actinide Elements for Use in Isotopic Power Generators (in French), *Bulletin d'Information Scientifique et Technique (Paris)*, No. 104, pp. 35-63 (May 1966) (English translation by R. G. Mansfield, ORNL-tr-1496).

‡D. E. Ferguson, Transuranium-Element Processing, *Isotopes and Radiation Technology*, 4(4): 321-324 (Summer 1967).

Section

IV

Isotopes and Radiation Technology

Process Radiation Development

Relation of Design Parameters, Plant Capacity, and Processing Costs in ^{60}Co Sterilization Plants*

By M. G. Brown†

Abstract: The main features of three sizes of ^{60}Co sterilization plants, designed to provide a complete range of capacities for radiation sterilization of medical products, are described. The smallest plant has a capacity of up to 50 thousand cu ft of medical products per year; the intermediate, up to 500 thousand cu ft; and the largest, over 1 million cu ft. The relations between capital costs, production rate, efficiency, and unit processing costs for each size of plant are discussed, and the method for selecting the best size of plant for a particular need is outlined.

The Commercial Products Group of Atomic Energy of Canada Limited (AECL) has developed designs for three sizes of ^{60}Co sterilization plants, which provide a complete range of capacities for the radiation sterilization of medical products. AECL completed its first ^{60}Co sterilization plant in 1964. This high-capacity plant—constructed for Ethicon Somerville Inc., at Somerville, N. J., United States—is still one of the most efficient plants in the world. Several other designs were developed to meet a number of industrial requirements, and now a complete range of sizes is available for industrial applications. The high-capacity design is similar to that of the original Ethicon plant at Somerville. The intermediate- and low-capacity plants incorporate many of the design features of the Ethicon plant but have some basic differences that result in lower capital costs while maintaining high radiation and plant efficiencies.‡

There are two types of low-capacity plants—manual and automatic. Plants of all three sizes comprise a ^{60}Co source and source-transfer mechanism, a source-storage unit, an irradiation cell (or biological shield), and a product conveyor or transfer mechanism.

Plant Design

Typical performance characteristics for the different size plants are shown in Table 1.

HIGH-CAPACITY PLANT

The high-capacity plant (Fig. 1) has a ^{60}Co source capacity up to 1.5 Mc. The basic source element used is the standard AECL "source pencil," which consists of sixteen 0.25-in.-diameter by 1-in.-long ^{60}Co slugs, doubly encapsulated in stainless-steel tubing sealed by welding. Pencils are loaded into frames, or modules, with 42 pencil positions. The modules are then assembled into a rack that can accommodate up to 20 modules in two rows of 10, one above the other. This arrangement permits installation of up to 1.5 Mc of ^{60}Co of 30 curies/g specific activity and allows adequate space for replenishment, necessitated by radioactive decay, without removing any activity.

The ^{60}Co source rack is raised from the source-storage pool by a pneumatic hoist, which returns the source to the pool automatically in case of power failure. The rack is guided by stainless-steel cables. The source and the mechanism are housed in an irradiation cell (biological shield) constructed of standard density (2.36 g/cm^3) concrete. The control area is equipped with a console, where machine controls and radiation-monitoring instruments are located. A safety interlock system is provided to ensure that

*Report AECL-2870, presented at IAEA Symposium on Radiosterilization of Medical Products, Budapest, June 5–9, 1967. Reprinted, with minor editorial changes, by permission of author.

†Atomic Energy of Canada Limited, Commercial Products Division, Ottawa.

‡Radiation efficiency [Canadian definition] is the ratio of the radiation absorbed by the product to the total radiation emitted by the radiation source. For a ^{60}Co plant, efficiencies up to 40% can be achieved, where efficiency is based on the minimum dose delivered. Plant efficiency is defined as the percentage of time the plant is available for use and is usually higher than 95% for a ^{60}Co plant.

Table 1 TYPICAL PERFORMANCE CHARACTERISTICS OF DIFFERENT-SIZE MEDICAL-PRODUCTS GAMMA-STERILIZATION PLANTS

Packing density, g/cm³	Ratio of max. dose to min.	Efficiency, *%		Processing rate† per kilocurie of ^{60}Co			
				Automatic		Batch	
		Automatic	Batch	cu ft/hr	m³/hr	cu ft/hr	m³/hr
High-Capacity Plant: Annual Production Rate‡ 1.04 cu ft/curie							
0.10	1.19	18.5		0.145	0.0040		
0.15	1.21	26.2		0.137	0.0038		
0.20	1.23	33.2		0.130	0.0036		
0.25	1.25	38.3		0.120	0.0033		
Intermediate-Capacity Plant: Annual Production Rate‡ 0.94 cu ft/curie							
0.10	1.30	17.9		0.134	0.0037		
0.15	1.31	25.3		0.127	0.0035		
0.20	1.33	31.3		0.118	0.0033		
0.25	1.35	36.0		0.108	0.0030		
Low-Capacity Plant: Annual Production Rate‡ 0.40 cu ft/curie Automatic, 0.32 cu ft/curie Batch							
0.10	1.27	6.6	6.2	0.0498	0.0014	0.0465	0.0013
0.15	1.28	9.9	9.2	0.0496	0.0014	0.0463	0.0013
0.20	1.28	13.1	12.2	0.0493	0.0014	0.0459	0.0013
0.25	1.29	16.1	15.0	0.0488	0.0013	0.0452	0.0012

*Based on source content and minimum dose.
†Based on minimum dose of 2.5 Mrads.
‡Based on 0.2-g/cm³ material and 8000 hr automatic (7000 hr in the case of the low-capacity batch) operation.

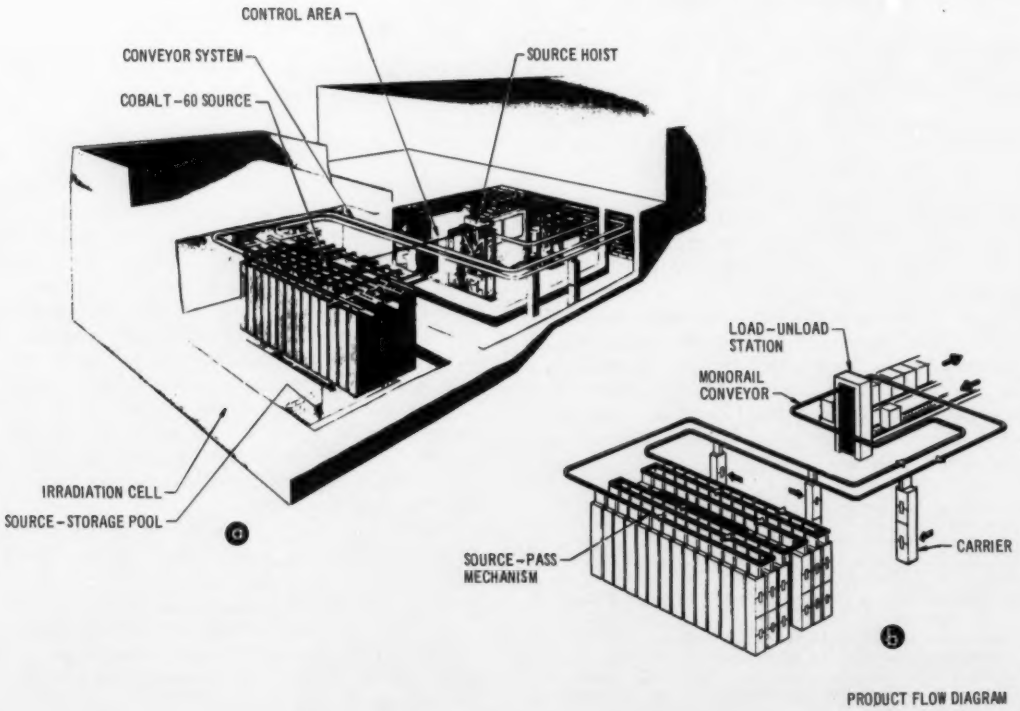


Fig. 1 (a) High-capacity plant and (b) its product flow diagram.

the operating staff will not be in the irradiation room or maze when the source is exposed.

The product to be sterilized is packed in standard size boxes, 12 by 15 by 21 in., which are carried along in pairs (taped together) by the conveyor system. At the load–unload station, these boxes are stacked automatically on carriers and carried through the entrance maze by a monorail conveyor. At the entrance to the irradiation room, the carriers are transferred to a source-pass mechanism, which consists typically of six monorail lines (three on each side of the ^{60}Co source rack) and several pneumatic transfer mechanisms, which move the carriers around the source.

The carriers travel along the six monorails in intermittent motion so that each box spends a fixed amount of time in each carrier position. Carriers arriving at the "receive" position are indexed by the pneumatic transfer mechanisms through all positions in the six rows and then are transferred to the outgoing monorail line at the discharge station. This arrangement results in a high absorption of radiation by the product with only a small variation in absorbed radiation dose in a lateral direction through the product box. The variation in absorbed dose in the vertical direction is decreased to an acceptable level by conveying the carton pairs through the irradiation cell first in the upper section of the carrier and then in the lower section. The transfer of vertical position takes place automatically at the load–unload station outside the irradiation cell. This arrangement results in an overall maximum-to-minimum absorbed dose ratio (overdosing ratio) of approximately 1.23 to 1 for any one product box with a density of 0.2 g/cm^3. Handling the boxes in pairs effectively doubles the maximum processing rate of the conveyor system, simplifies the design, and decreases the cost of any automatic storage system for unattended operation. However, the dose ratio can be improved further by handling boxes individually instead of in pairs and by conveying each box through the irradiation cell four times.

The number of product rows on each side of the source is optimized for a given source size. For plants with a low or medium initial rate of production, the irradiator may be built with only one layer on each side of the source, with provision for the addition of extra layers when the production rate increases. The extra layers can be added within a normal plant shutdown period. The use of fewer product layers with a small source reduces the product holdup in the irradiator, which may be important in some plants.

INTERMEDIATE-CAPACITY PLANT

The basic components of the intermediate-capacity plant (Fig. 2) are the same as those of the high-capacity plant—a ^{60}Co source and source-transfer mechanism, a storage pool, an irradiation cell, and a product conveyor system. The source and source-transfer mechanism are similar to the units described for the high-capacity plant, as is the irradiation cell. The same source pencils and modules are used, but the source rack can accommodate only six modules. Up to 500 thousand curies of ^{60}Co of 30 curies/g specific activity can be installed, and there is adequate space for replenishment without removing any activity.

Standard-size (18 by 18 by 18 in.) boxes of medical supplies are taped together in pairs and carried along by the supply conveyor system. At the load–unload station, these boxes are transferred automatically to the upper half of the product carrier and are carried through the entrance maze by a monorail conveyor to the source-pass mechanism (Fig. 3). This mechanism comprises two levels of box-support trays, pushers, and end transfers and an elevator. The incoming product boxes are pushed onto the upper-level tray and are conveyed about the source by the pneumatic transfer devices and an elevator. At the same time, the sterilized boxes are pushed onto the lower tray of the product carrier, which returns through the maze and transfers the boxes to the discharge conveyor.

LOW-CAPACITY PLANT

There are two types of low-capacity plant—batch and automatic—and both types may be designed with water-pool source storage or with dry source storage using a transportable shipping container. The latter is suitable where access to a hot cell for source replenishment is possible. The main advantage of these plants is low capital cost. Figure 3 shows a batch plant and Fig. 4 an automatic plant.

Both batch and automatic plants include a 9-in.-diameter by 17-in.-high annular source arrangement built from standard AECL ^{60}Co source pencils. Up to 50 thousand curies of ^{60}Co may be installed in the dry-storage unit and 100 thousand or more curies in the pool-storage unit. A "fail-safe" pneumatic hoist is used to move the source between the storage and exposure positions. The two plants have similar concrete irradiation cells and simple control and safety systems.

The batch-plant source-pass equipment consists of seven square platforms held in an eight-space lattice. Three standard-size (16 by 16 by 16 in.) product boxes are stacked on each platform, and the mechanism conveys the boxes around the source. During the 20- to 30-hr irradiation, the boxes are rearranged on the tiers to ensure dose uniformity.

The automatic-plant conveying system carries the same size product boxes from the supply conveyor through the irradiation cell and returns them to the discharge conveyor. Incoming boxes are transferred by a pneumatic device to the product carrier and carried to the source-pass mechanism by a monorail

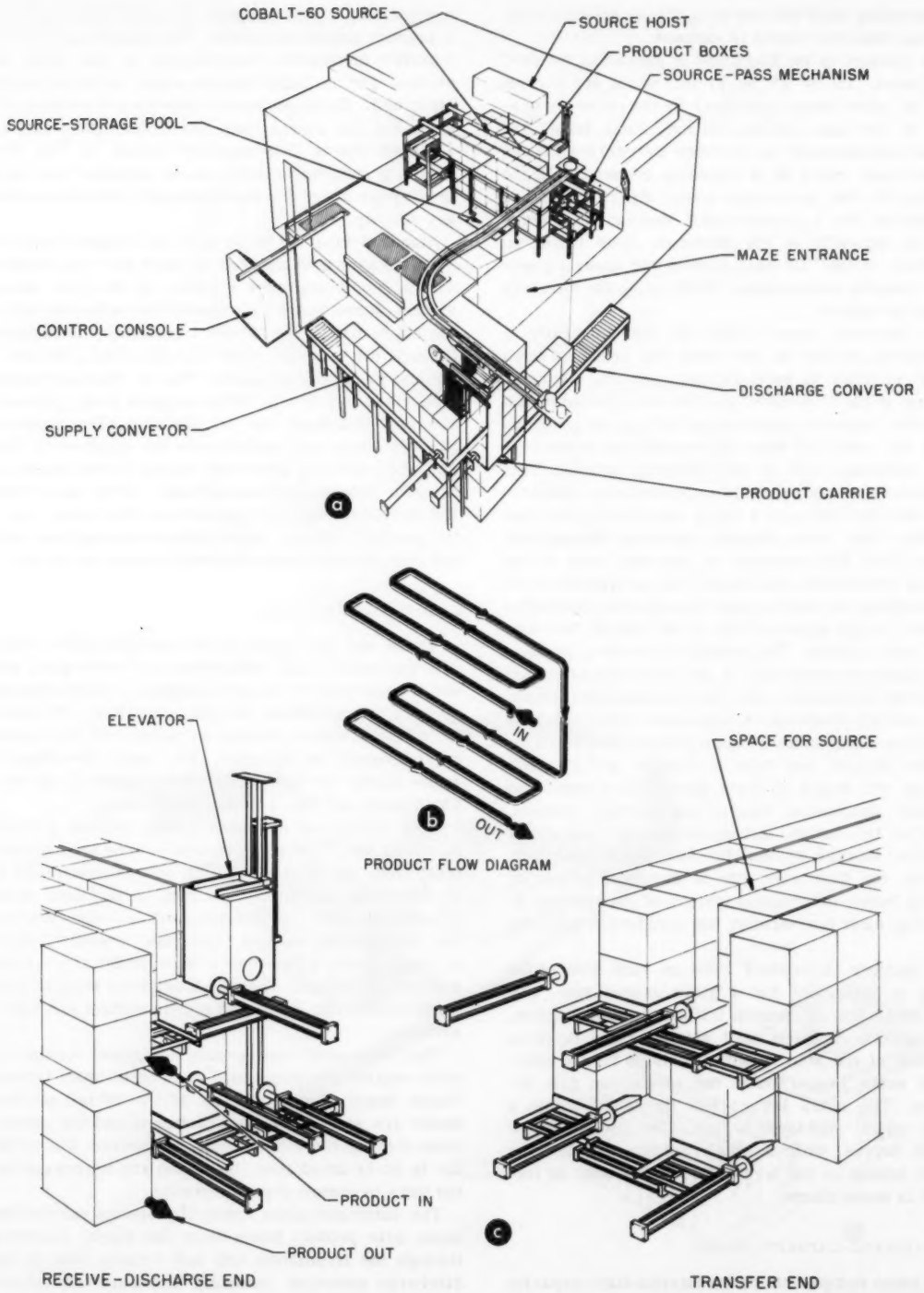


Fig. 2 (a) Intermediate-capacity plant, (b) product flow diagram, and (c) source-pass mechanism.

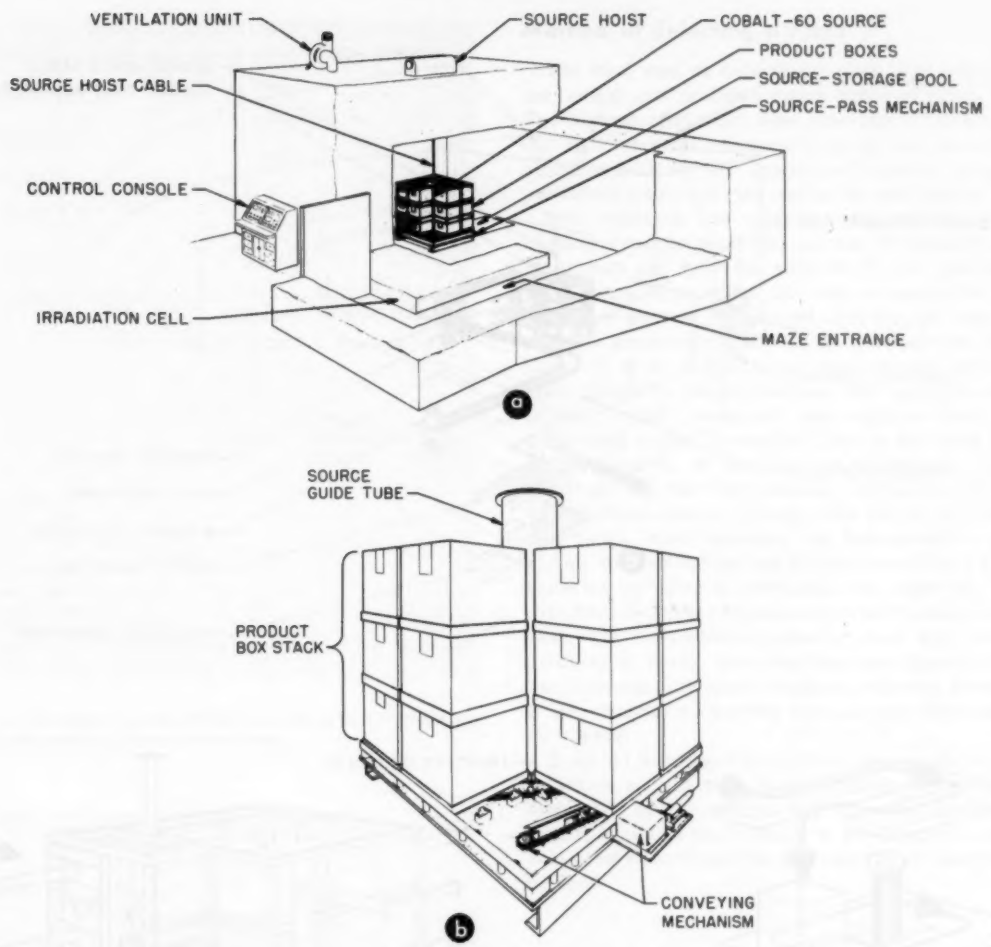


Fig. 3 (a) Low-capacity batch plant and (b) its source-pass mechanism.

conveyor. At the source-pass transfer point, an incoming box is pneumatically transferred to the source-pass mechanism, which conveys the product boxes around the ^{60}Co source automatically. Pneumatic transfer and elevating devices move the product boxes through 24 lattice positions on three levels of box support trays. At the same time, the irradiated box is discharged from the source-pass mechanism to the product carrier, carried back through the maze, and transferred to the discharge conveyor.

Plant and Processing Costs

Figure 5 indicates the capital and annual cobalt costs for the four plants, with cobalt sources of from 10 to 1400 kilocuries; Fig. 6 indicates costs per cubic foot of product irradiated for production rates of from 8 thousand to more than 1 million cu ft/year. We see that the minimum capital cost for a low-capacity batch plant with a 25-kilocurie source is about $100 thousand ($73,000 + $22,000), and the processing cost with this plant investment is $2.75 per cubic foot for a production rate of 8 thousand cu ft/year. For a high-capacity plant with a 1200-kilocurie source, the capital cost is about $1 million, or 10 times the minimum for the low-capacity plant, and the processing cost is less than 20 cents per cubic foot for a production rate of 1.2 million cu ft/year. For production rates up to 50 thousand cu ft/year, unit costs range from $3 to $1, depending on the quantity processed; they drop sharply, to about 50 cents, for a rate of 200 thousand cu ft/year; and then decline slowly to less than 20 cents for annual production rates greater than 800 thousand cu ft.

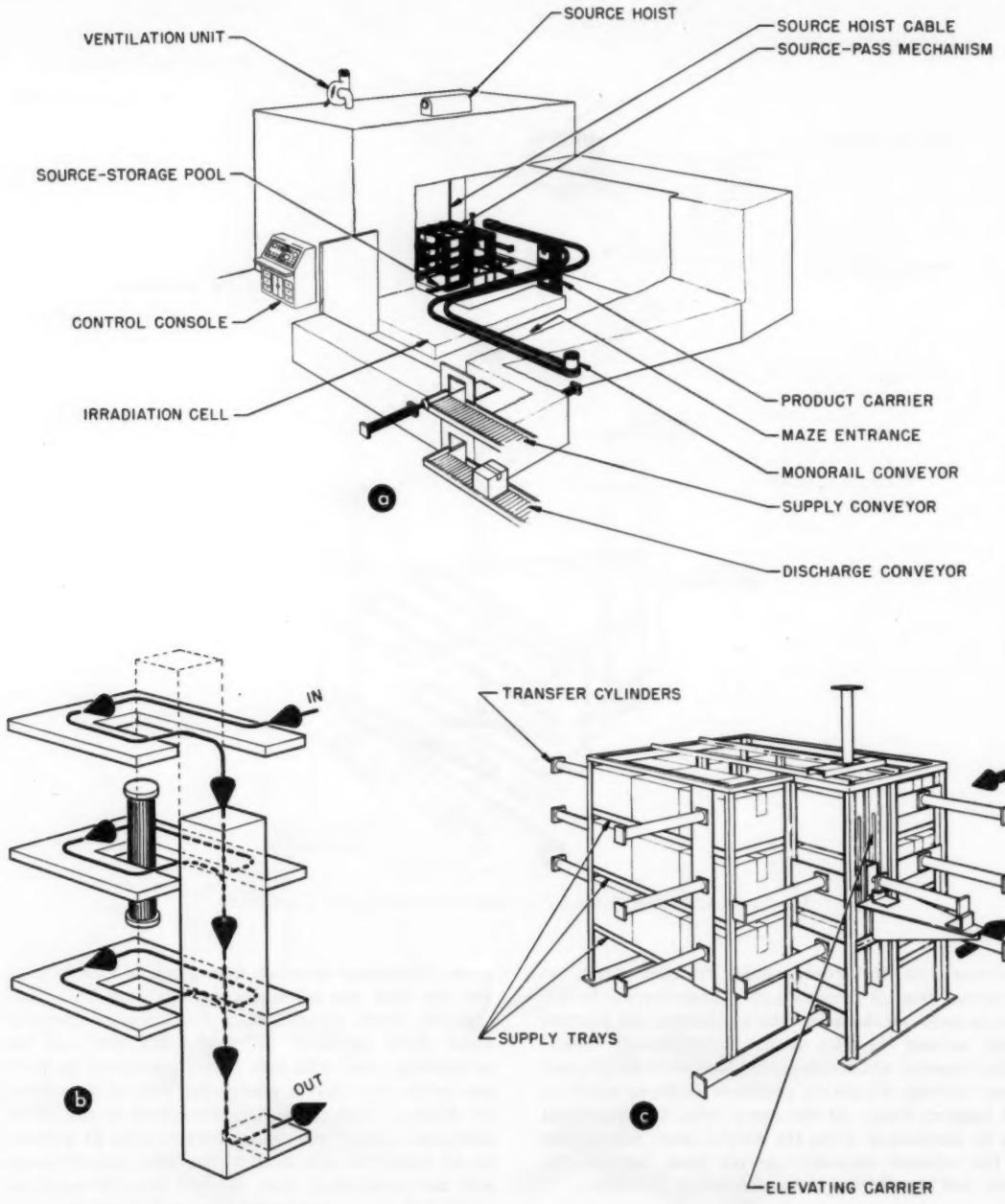
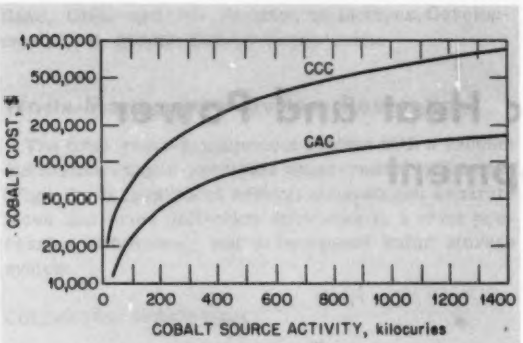


Fig. 4 (a) Low-capacity automatic plant, (b) product flow diagram, and (c) source-pass mechanism.



	Low capacity		Intermediate capacity	High capacity
	Batch	Automatic		
Total capital = CCC + machinery and cell	$73,000	$101,000	$145,000	$265,000
Total annual = CAC + depreciation, interest, services, and labor	$16,500	$ 21,000	$ 32,500	$ 56,000

Fig. 5 Cobalt-60 capital (CCC) and annual (CAC) costs for medical-products-sterilization plant.

Method of Selecting a Plant

The first step in selecting a plant is to determine the initial and probable future rates of production. The highest production rate expected will determine the type of plant required. A plant may have to be operated initially at a fraction of capacity if higher production rates are expected in the near future.

Both types of low-capacity plants and the intermediate-capacity plant will process 20 thousand cu ft of product per year for about $1.50 per cubic foot. The choice of plant for this rate of production will depend on whether 20 thousand cu ft is a low, average, or high processing rate for the planned life of the plant. If it is a high value, the low-capacity batch plant should be chosen because this plant offers the lowest capital investment and slightly lower unit processing costs. Production costs in the range from 25 thousand to 50 thousand cu ft/year are almost identical for the low-capacity automatic and the intermediate-capacity plants. The choice of plant in this range must therefore be determined by estimating the probability that 50 thousand cu ft is a high value for the ultimate production rate expected.

In the range from 50 thousand to 300 thousand cu ft/year, the intermediate-capacity plant will process products at lower costs than the high-capacity plant. The high-capacity plant should be selected, however, if the ultimate production rate exceeds 500 thousand cu ft/year.

It should be pointed out that the above guide to plant selection may have to be modified due to factors not mentioned in this article. For example, the limit on capital available might result in the choice of a lower cost plant even though the unit costs of production are higher. (MG)

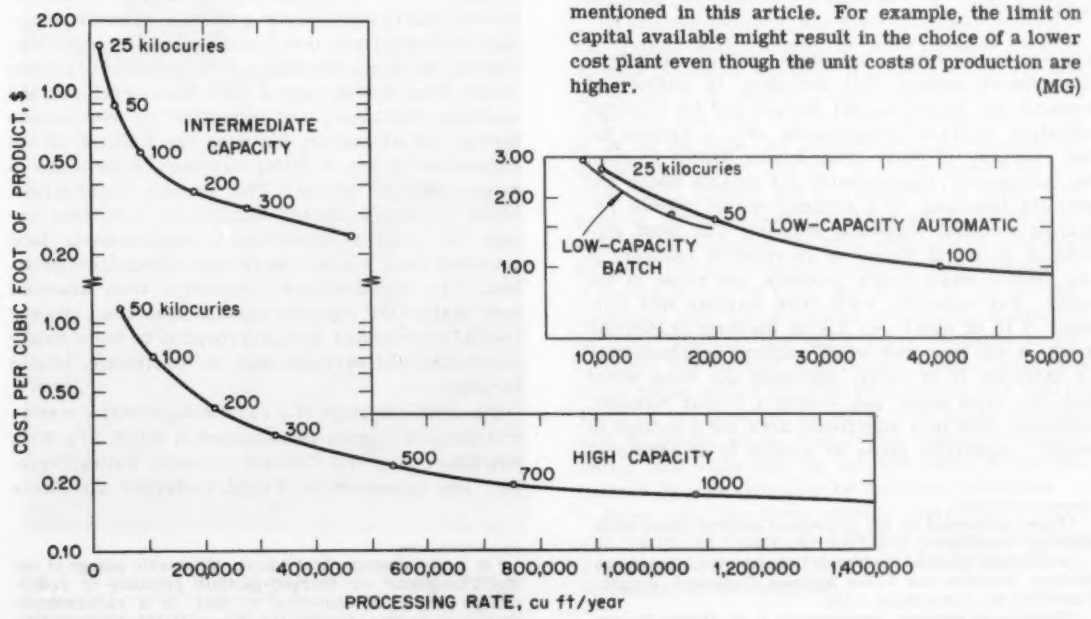


Fig. 6 Unit processing costs for ^{60}Co medical-products-sterilization plants of different capacities.

Section V

Isotopes and Radiation Technology

Isotopic Heat and Power Development

Integrated Waste-Management System with a Vacuum Distillation–Vapor Pyrolysis Radiosotope-Fueled Water-Reclamation Function*

By C. A. Metzger,† A. B. Hearld,† B. J. Reynolds,† R. Shivers,‡ and R. W. Murray§

Abstract: The first prototype of a total waste-management system for an aerospace life-support system with the water-recovery function thermally powered by a radioisotope is reported. Five hundred grams of ^{238}Pu [0.56 watt(t)/g] was used to supply a total of 280 watts(t) from four 70-watt(t) heat sources. The small electric-power requirement for the pumps and valves was met by small external supplies. The prototype was designed and tested to supply two men with water for a 30-day mission, and it recovered approximately 0.75 lb of water per hour from waste liquids.

Long-term aerospace missions will necessitate a life-support system that includes, in addition to systems for environmental control and for catalytic oxidation of trace contaminants, etc., a system for the recovery of water from human wastes to meet the astronauts' requirements for potable water and permits launching of a minimal-weight vehicle requiring minimal logistical support. The most significant saving in weight is provided by recovery of the crew's waste-water products and reuse of the water. For example, each crew member will consume 6 lb of water per day in the form of food and drink. A six-man crew would consume approximately 13 thousand lb of water, excluding any wash water that the crew might use, during a 1-year mission. Obviously this is a significant area for a savings in weight, especially since 20 million lb of additional booster thrust would be required to launch the 13 thousand lb of water on an interplanetary mission.

Design criterions for a waste-management system dictate minimum or maximum standards for (1) bacteria count, (2) potability, (3) expendables, (4) total energy, (5) weight and volume, (6) maintenance, and (7) reliability. Numerous processes and techniques—vapor compression, vacuum distillation, vacuum distillation–vapor pyrolysis, reverse osmosis, electrodialysis, membrane permeation, air evaporation, and dehumidification—for recovery of potable water from human wastes have been developed and systems fabricated and subjected to comparison testing and evaluation, but none has fulfilled all the requirements for a water-recovery system for a manned space vehicle. The vacuum distillation–vapor pyrolysis process fulfilled all criterions except the energy requirement. Radioisotopic heat sources¶ have solved the energy-requirement problem. The radioisotopes provided a truly constant heat output for constant system operation, and the radioisotope-heated system proved to be more easily maintained and serviced than an electrically heated version.

The first prototype of a radioisotope-heated waste-management system is discussed in detail. The work was done by General Electric Company, Valley Forge, Pa., and sponsored by Wright-Patterson Air Force

*Paper presented at the Aerospace Medical Association Meeting, Washington, D. C., Apr. 13, 1967.

†Aerospace Medical Research Laboratories, Aerospace Medical Division, Air Force Systems Command, Wright-Patterson Air Force Base, Ohio.

‡Division of Isotopes Development, U. S. Atomic Energy Commission, Washington, D. C.

§General Electric Company, Valley Forge, Pa.

¶In a radioisotopic heat source, the kinetic energy of the electromagnetic or charged-particle products of radioisotope decay is converted to heat. In a radioisotopic electrical source, the kinetic energy is transformed into electricity either directly, or indirectly through a preliminary conversion to heat.

Base, Ohio, and the Division of Isotopes Development, U. S. Atomic Energy Commission.

Waste-Management System Description

The total waste-management system with a vacuum distillation–vapor pyrolysis water-recovery function (Figs. 1–3) consists of several subsystems: separate feces and urine collection subsystems, a urine processing subsystem, and a recovered-water storage system.

COLLECTION SUBSYSTEMS

Fecal-Collection Waste-Management Subsystem. The collection and storage of biological wastes is a psychological and sanitary problem that is amplified by the restrictions and requirements of space flight. Since feces contain large numbers of bacteria and are generally very adhesive, it is of prime concern to minimize handling and crew exposure to this material. The system utilizes a technique of waste management similar to earth procedures; i.e., the man defecates, wipes with tissue, and then has no further exposure to the waste material or need to transport bags containing the material. The collector (Fig. 4) is designed for four men on a 30-day mission in an aerospace environment.

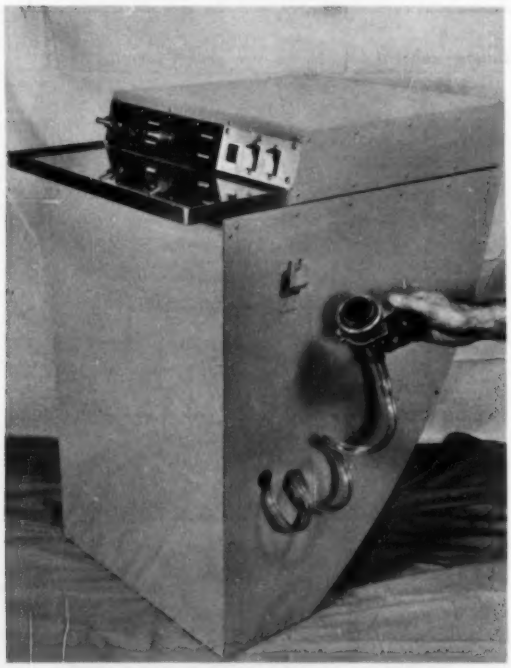


Fig. 1 Exterior view of 30-in.-high by 22-in.-wide by 30-in.-deep prototype of water-recovery function of waste-management system. The curvature of the back wall is designed to conform to the shape of a spacecraft wall. The device on the side of the system is the urinal.

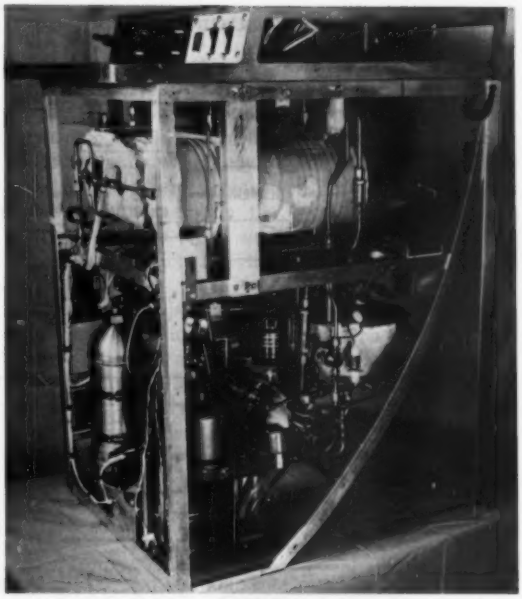


Fig. 2 Interior view of prototype of water-recovery function of waste-management system.

To use the collector, the crewman seats himself in the flexible seat designed to index the user over the storage container opening. If desired, an air jet may be activated to assume proper positioning. The user defecates normally and the stool is transported (Fig. 5) by an air flow (for zero-gravity operation) to the storage container. The air is drawn through the unit centrifugal blower and is vented to the cabin after passing through a bacterial filter and activated charcoal. In the container the stool impinges on a rotating impeller, is centrifugally slung outward, shredded by a series of rotating tines, and spread on the storage-container wall. Dense packing of the feces is achieved by this method, and each layer of feces is sufficiently thin so that it is rapidly vacuum dried. The toilet tissue enters the storage container in a similar manner, but is either shredded or slides up the tines and is stored whole. The Teflon coating of the impeller and tines and the outward angle of the tines prevent impeller clogging by the tissue and feces.

After use, the feces storage container is sealed and vented to vacuum by a bacterial filter. The feces are vacuum dried to provide a bacteriostatic environment in the container.

Urine-Collection Subsystem of Water-Recovery Function. The collection subsystem consists of a

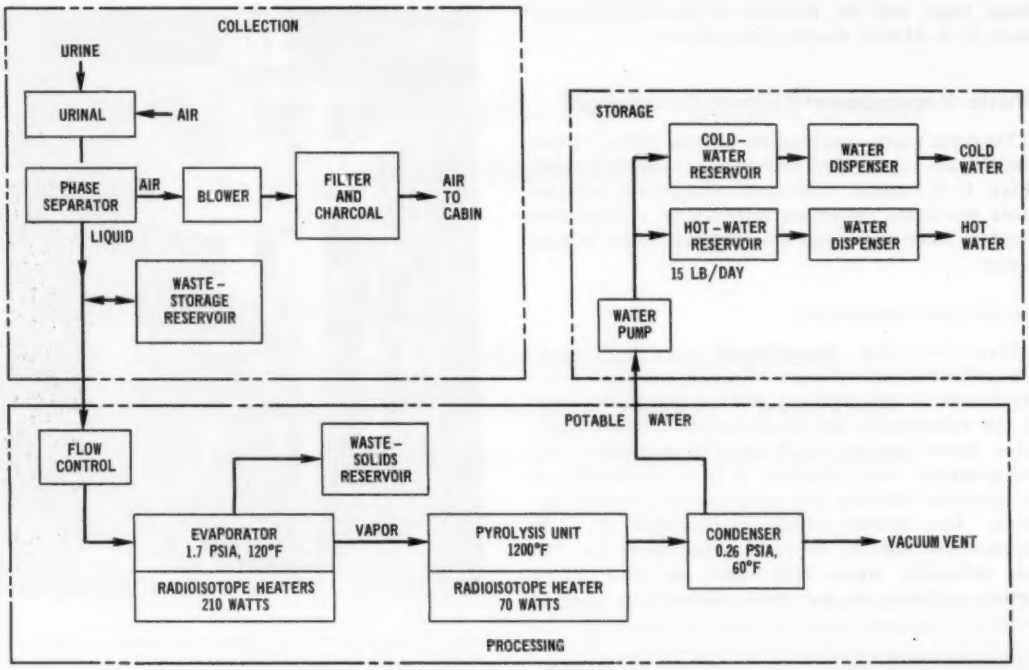


Fig. 3 Diagram of radioisotope-heated vacuum distillation–vapor pyrolysis water-recovery function.

Fig. 4 Fecal waste collector.

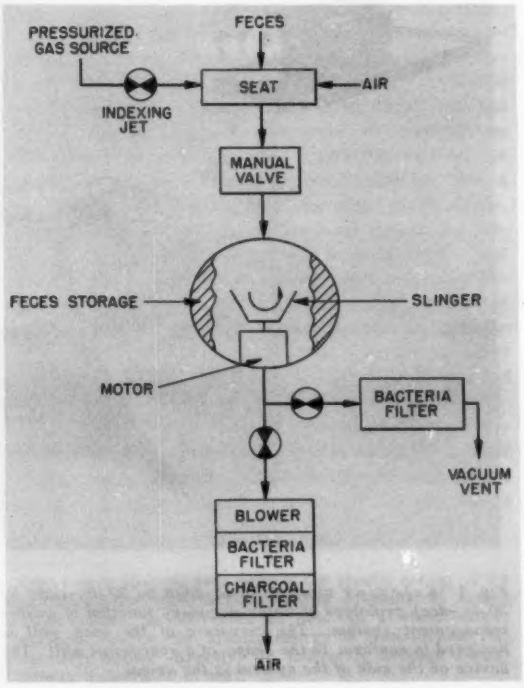


Fig. 5 Block diagram of fecal-collection waste-management subsystem.

urinal, phase separator, waste-liquid storage reservoir, blower, air filter, and controls.

The user micturates into the urinal after starting the phase separator and blower. The urinal has an adjustable orifice that provides a positive and gentle seal. For zero-gravity operation the urine is conveyed by a cabin air stream (which enters through the urinal) to the phase separator where the air is centrifugally separated, drawn through a blower, some bacteria charcoal, and a filter, and vented to the cabin. The urine is pumped to a storage reservoir from which it is metered into the urine processing subsystem.

URINE PROCESSING SUBSYSTEM OF WATER-RECOVERY FUNCTION

This processing subsystem includes an evaporator unit, a pyrolysis unit, a condenser unit, and pumping systems.

Evaporator Unit. The evaporator unit (Fig. 6) consists of a fill control, the evaporator, and a phase separator.

The evaporator is filled as required to maintain a certain liquid level, which is accomplished by an ultrasonic level sensor in the evaporator. Liquid is added from the waste-liquid reservoir by electrically opening two series-redundant solenoid valves as necessary to maintain the level. Originally, conductivity probes were used for level sensing, but these were short circuited by solid accumulations on the probes.

The urine evaporator is a vessel containing ports for the admission of urine and the discharge of steam. The urine input to the evaporator contains approximately 5% solids, which are separated from the liquid (vapor) by the distillation process. The solids are then centrifugally conveyed to the storage reservoir by the impeller in the evaporator. Heat, to 120°F, is provided by three 70-watt(t) ^{238}Pu heat

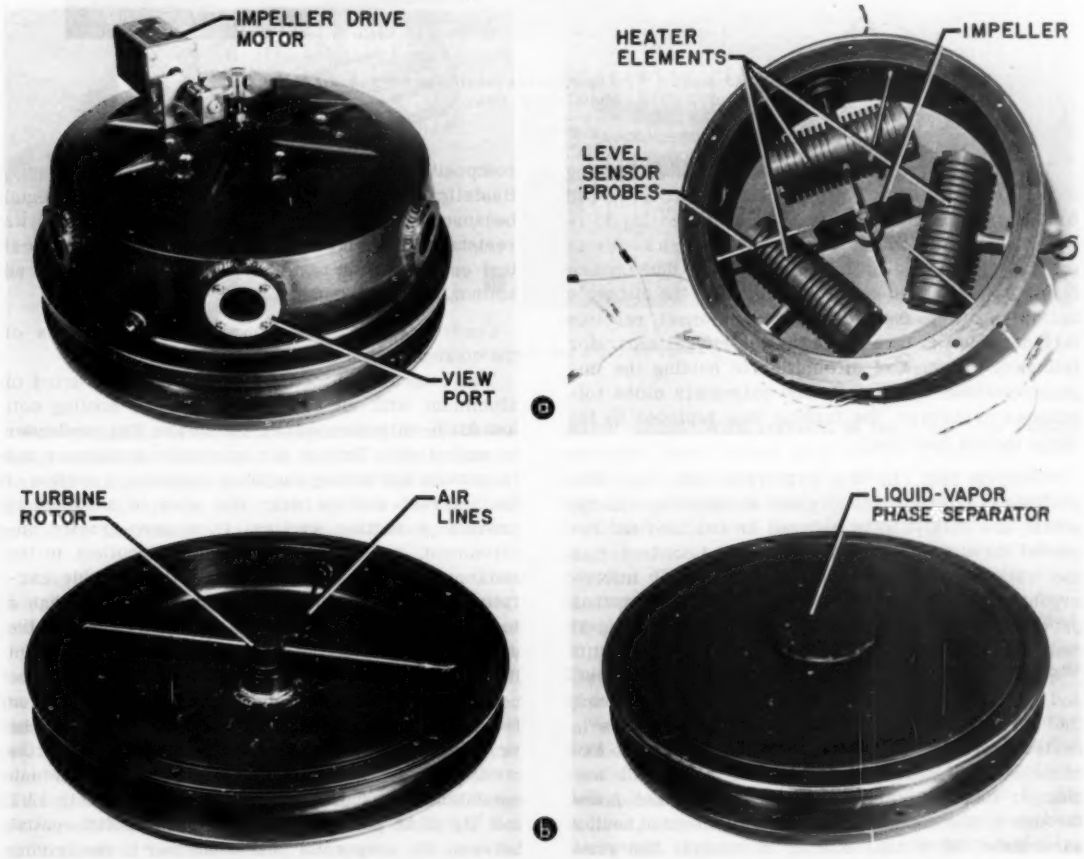


Fig. 6 (a) Exterior and (b) interior views of the evaporator unit of the water-recovery function.

sources (Fig. 7). The liquid boils at reduced pressure, i.e., 87.5 mm Hg (1.7 psia). A bladed rotor driven at 60 rpm by the evaporator motor and gearbox through a Quad-Ring shaft seal provides the gravitational field necessary for convective nucleate boiling in a zero-gravity environment. The evaporator is constructed of 6061-T6 aluminum, coated with Tufram for corrosion resistance.

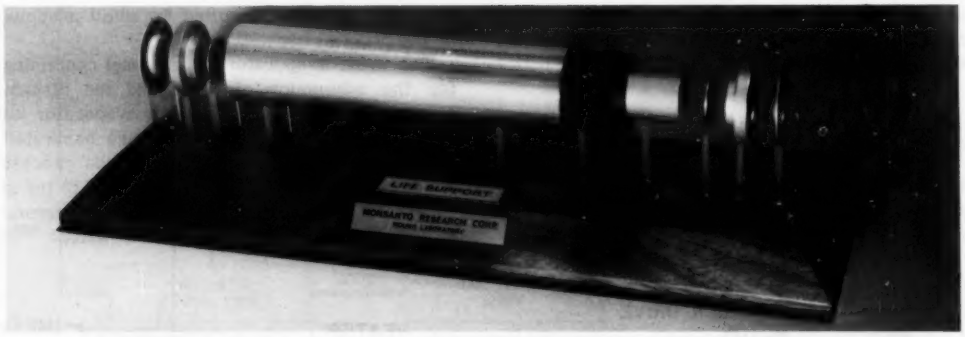


Fig. 7 The 70-watt(t) ^{238}Pu heat source fabricated by Mound Laboratory, Monsanto Research Corp., Miamisburg, Ohio.

The phase separator consists of a superheating exchanger and a baffled exit. The steam is passed through the heat exchanger to ensure that liquid is not carried with the steam flow. The original design of the phase separator had a slinger and turbine drive, so that the steam flowing through the slinger's hollow shaft passed through a pinwheel reaction turbine which in turn drove the centrifugal separator (slinger). Because of difficulties in holding the impeller-turbine bearings to the extremely close tolerances necessary, the turbine was replaced by the static device described.

Pyrolysis Unit. In the pyrolysis unit, the impurities — ammonia and organic substances — in the steam are catalytically reduced or oxidized and removed through the vacuum vent in the condenser as the water vapors are liquefied. Also, any microorganisms in the vapor stream are incinerated, thus providing a sterile vapor. The pyrolysis unit (Fig. 8) consists of a counterflow heat exchanger, a 70-watt(t) ^{238}Pu heat source, ruthenium or rhodium catalyst, and an evacuated insulation jacket. Steam entering the pyrolysis unit through the exchanger tubes is heated to 1200° F by the steam exiting from the exchanger. The incoming steam flows through the exchanger tubes, exits from the exchanger, and flows through a tube containing the centrally located heater surrounded by a thin coating of catalyst that performs the function of assisting the heat transfer to the steam as well as acting as a catalyst to complete the pyrolysis process. Upon leaving the heater, the steam enters the catalyst bed and turns and flows back through another portion of the catalyst bed to the counterflow exchanger and out, giving up some of its heat to the incoming steam.

Early in the design phase of the program, it was believed that a steam temperature of 1800° F was required to effect sufficiently complete ammonia decomposition with a platinum catalyst. Accordingly, Hastelloy X was chosen for the fabrication material because of its strength at that temperature and its resistance to corrosion in the common metallurgical test environments; tests have confirmed its use in an ammoniacal environment.

Condenser Unit. The condenser unit consists of the condenser and the condenser-level control.

The conically shaped condenser is constructed of aluminum with an externally wrapped cooling coil and foam–silicone–rubber insulation. The condenser is coated with Tufram for corrosion resistance and to provide nonwetting surfaces; however, a section of the internal surface near the apex is anodized to provide a wetting surface. In a zero-gravity environment, the condensed water will collect in the minimum-energy configuration at the wettable surface of the apex. Internally, the condenser has a baffle at the steam inlet to deflect the steam over the cool wall for better heat transfer. The vacuum vent line that controls the system pressure enters the condenser in the steam line and the heat transferred from the steam eliminates the possibility of the vented gas freezing in the vent line. The vapor is condensed at approximately 15.6°C (60° F), which establishes a vapor pressure of approximately 13.2 mm Hg (0.26 psia). The vapor-pressure differential between the evaporator and condenser is the driving force that causes the vapor to flow through the subsystem.

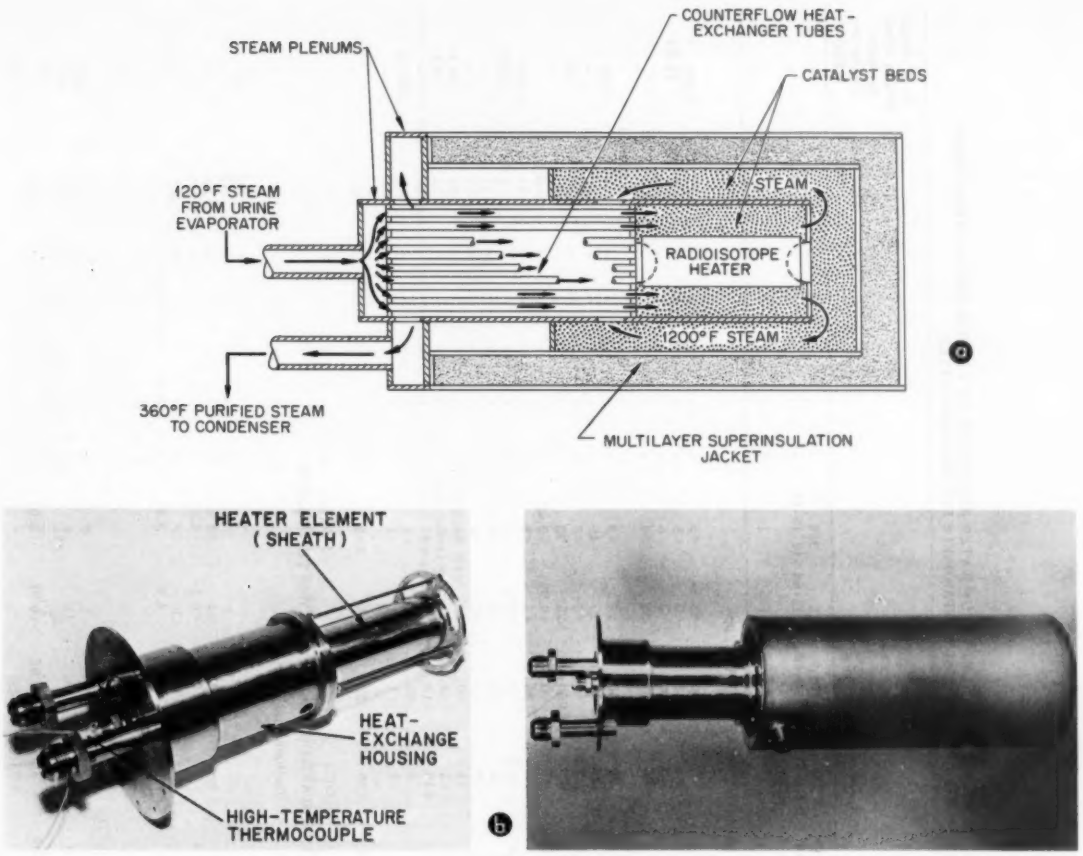


Fig. 8 (a) Sectional view and (b) interior and exterior views of the pyrolysis unit.

The condenser-level control is an electrical timer, which activates the pump at a rate slightly faster than normally required. This ensures that the water level will always be controlled at a low level, thus providing a constantly large condenser surface area.

Pumping Systems for Recovered Water. The pumping systems consist of a pump for potable water and one for solids.

The pump for potable water draws the water from the apex of the condenser. The pneumatically powered positive-displacement piston pump produces a suction, so that the water flows through a check valve and into the cylinder of the pump. Gaseous oxygen then drives the piston forward, forcing the water through another check valve and into the water-storage reservoir. The oxygen is then vented to the cabin where it is consumed by the crew.

The initial design for the solids pump admitted a slurry of urine and urine solids to a cam-actuated solids pump. With rotation of the pump, the piston advanced from bottom dead center toward top dead center, first squeezing the slurry and permitting liquid to permeate through a porous plug in the piston head and get behind the piston, simultaneously trapping the solids ahead of the piston. Further rotation exposed the trapped solids ahead of the piston to a discharge port leading to the solids-storage reservoir. Rotation past the discharge port then resulted in retraction of the piston and backflushing of the fluid through the porous plug, along with admission of additional slurry for the next pumping cycle.

Wear resulting from unanticipated, extremely abrasive action of the solids in the urine suggested the need for a different mechanism of solids separation. Accordingly, the port which previously led to the solids pump has been directly connected to the solids reservoir, separation now being effected inertially, utilizing the centrifugal field established by the evaporator rotor. Also, separation is accomplished

Table 1 RESULTS OF ANALYSES OF SAMPLES OF SIMULATED URINE AND OF 15 SAMPLES OF WATER RECOVERED BY VACUUM DISTILLATION–VAPOR PYROLYSIS PROCESS AND COMPARISON WITH PUBLIC HEALTH STANDARDS FOR DRINKING WATER

	Recovered water															Simulated urine	U. S. Public Health drinking water standards (1962)
Sample No. (TC)	313	314	315	316	317	318	319	320	321	322	-	-	-	-	-	296	-
Date	11-18-66	11-18-66	11-18-66	12-6-66	12-6-66	12-3-66, 12-4-66	12-5-66, 12-6-66	12-13-66	12-15-66	12-15-66	-	-	-	-	-	9-7-66	-
Heat source	*	*	*	†	†	†	†	†	†	†	‡	‡	‡	‡	‡	-	-
SPECTROGRAPHIC DATA, ppb																	
Zinc	36	130	26	200	250	14	38	35	63	<5	-	-	-	-	-	<27	5,000
Cadmium	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	-	-	-	-	-	<27	10 R§
Boron	18	30	18	8	<3	14	<5	7	7	<5	-	-	-	-	-	325	-
Phosphorus	<20	105	<20	90	60	<25	<25	55	55	30	-	-	-	-	-	Major constituent	-
Iron	5	73	7	75	75	26	20	40	25	9	-	-	-	-	-	14	300
Molybdenum	¶	¶	¶	¶	¶	¶	<10	<10	<10	<10	-	-	-	-	-	<54	-
Manganese	<3	9	<3	8.5	<2.5	<2.5	3.5	<3	<3	<3	-	-	-	-	-	<14	50
Aluminum	1,375	>1,500	1,500	2,250	1,500	1,000	465	1,000	525	70	-	-	-	-	-	<54	-
Beryllium	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	-	-	-	-	-	<0.14	-
Copper	500	500	350	225	175	6	7	25	<3	<3	-	-	-	-	-	<14	1,000
Silver	<0.5	5	<0.5	<0.5	<0.5	<0.5	<0.5	4	<0.5	3	-	-	-	-	-	<2.7	50 R
Nickel	<5	20	<5	16	<5	<5	<5	<5	<5	<5	-	-	-	-	-	<27	-
Cobalt	<5	<5	<5	<5	<5	<5	<5	<5	<5	<5	-	-	-	-	-	<27	-
Lead	<10	<10	<10	45	<10	15	15	40	<10	30	-	-	-	-	-	<54	50 R
Chromium	<3	17	<3	21	10	<5	<5	<3	<3	<3	-	-	-	-	-	<14	50 R
Vanadium	<10	<10	<10	<10	<10	<10	<10	<10	<10	<10	-	-	-	-	-	<54	-
Barium	4	12	4	5	14	1	4	80	<1	<1	-	-	-	-	-	<3	1,000 R
Strontium	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	-	-	-	-	-	19	-
Mercury	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Arsenic	<25	<25	<25	<25	<25	<50	<50	<25	<25	<25	-	-	-	-	-	-	-
U. S. PUBLIC HEALTH STANDARD WATER ANALYSIS TECHNIQUES																	
Cations, mg/liter																	
Calcium	<1	<1	<1	-	-	-	-	-	-	-	-	-	-	-	-	52	-
Magnesium	<1	<0.2	<1	-	-	-	-	-	-	-	-	-	-	-	-	102	-
Sodium	<1	1	<1	5	3.1	<1	4	<1	<1	<1	-	-	-	-	-	2,200	-
Potassium	<0.5	<0.5	<0.5	3.1	3.1	0.4	1	<0.1	<0.1	<0.1	-	-	-	-	-	1,700	-
Arsenic	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	0.05 R
Ammonia	87	250	75	10	6.3	17	8	1,500	4.2	3.0	5	3.5	0.03	0.03	0.4	-	-
Anions, mg/liter																	
Sulfate	45	60	35	12	9	11	21	25	<1	7	8	-	-	-	0	900	250
Chloride	9	250	7.5	48	36	6.5	6.5	18	11	4	5.5	38	12	3	3.5	3,700	250
Nitrate	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	10	45
Total phosphate	0.06	0.06	-	0.28	0.28	0.06	0.35	0.02	<0.01	<0.01	-	-	-	-	-	1,400	-
ABS**	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	0.5

OTHER TESTS

pH	9.2	9.2	9.1	4.1	4.4	8.9	8.2	9.7	4.0	8.3	3.8	4.2	3.9	4.5	5.3	-	-
Conductivity, μmhos/cm	270	1,570	260	205	165	175	170	2,900	77	116	120	130	100	25	14	-	-
Turbidity, Jackson units	-	-	-	0	0	0	0	-	-	-	-	-	-	-	-	-	-
Color	-	-	-	0	0	0	0	-	-	-	-	-	-	-	-	-	15
Odor	-	-	-	-	-	Slight urine	Slight urine	-	-	-	0	0	0	0	0	-	3
Total hardness, mg $CaCO_3$/liter	<1	<1	<1	14	5	6	6	<1	<1	<1	-	-	-	-	-	550	-
Total alkalinity, mg $CaCO_3$/liter	298	1,300	356	0	0	226	86	6,200	38	52	0	0	0	1	6	92	-
Total solids, mg/liter	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	500
Chemical O_2 demand, mg/liter	-	-	-	-	-	-	-	-	-	-	18	144	40	16	23	10,000	-
Urea, mg/liter	88	<0.1	<0.1	10	10	<0.1	<0.1	80	<0.1	<0.1	-	-	-	-	-	3,000	-
Total plate count	-	-	-	-	TMC††	-	0/ml††	-	-	<1/ml††	0	37	0	0	0	-	-
Total carbon, mg/liter	710	25	2,800	135	2,000	5.2	4	45	12	5	-	-	-	-	-	6,300	-

*Electrically heated pyrolysis and evaporator units.
†Radioisotopically heated pyrolysis and evaporator units.
‡Electrically heated pyrolysis unit and waste heat liquid for evaporator.
§Rejection: a maximum value that if exceeded means rejection; the other values are less critical.
¶Aluminum interference.
**U. S. Public Health standard tests for biodegradables, for which there is no need since no washing powders are added to the system.
††Agar pour plate technique.

without the need for the separate solids pump, with the slight penalty of incomplete recovery of liquid.

STORAGE SUBSYSTEM FOR RECOVERED WATER

The storage subsystem stores, heats or cools, and dispenses the potable water. The water is pumped through check valves into three storage reservoirs each with a capacity of 5 lb of water, where the pressure is 23.5 psig. Two of the reservoirs are cooled by a liquid coolant and one is heated by the steam exiting from the pyrolysis unit. To ensure filling of the hot-water reservoir and thus an ample supply of hot water, the hot-water reservoir check valve is set at 0.5 psi differential pressure and the cold-water reservoirs check valves at 4.0 psi differential pressure. The water is dispensed by the timed actuation of the water-dispenser solenoid valves; a variable time control holds the valve open until a calibrated amount of water is dispensed. Push buttons on the console permit dispensing of hot or cold water in 1- to 6-oz quantities. For example, if 5 oz of hot water is required to reconstitute a dried food, the user sets the selector switch to "5," attaches his food bag to the hot-water dispenser, and presses the hot-water switch.

Potability Meter

The potability meter indicates by a "go" or "no-go" signal if a water sample fails or meets the standards: *p*H between 5.5 and 8.0, specific conductivity not greater than 500 μmhos/cm, and for lack of other measurable electrolytes, less than 450 ppm chlorides and less than 2.5 ppm fluorides. Hopefully, a meter can be developed with analytical systems for carbon, odor, and color, as well as *p*H and conductivity. (The U. S. Public Health drinking water standards are presented in Table 1, pages 248-249, for comparison.)

The potability meter is an all-electronic analytical system based on the response of specific-ion electrodes. All system components are contained in a lightweight, portable, metal instrument case with overall dimensions of approximately 5 by 10 by 8 in. (Fig. 9). Weight is approximately 5 lb. The interior, which is accessible by a hinged, latched top, is divided into two compartments: one compartment contains the sampling manifold with mounted electrodes and necessary valving and the other contains all electronic components.

Red and green lights for "go" and "no-go" readout, a rotary switch for measurement selection, and valves and tubing connections for the manifold are mounted on the front panel of the instrument. All meter components were selected to give maximum ruggedness compatible with simplicity and ease of adjustment.

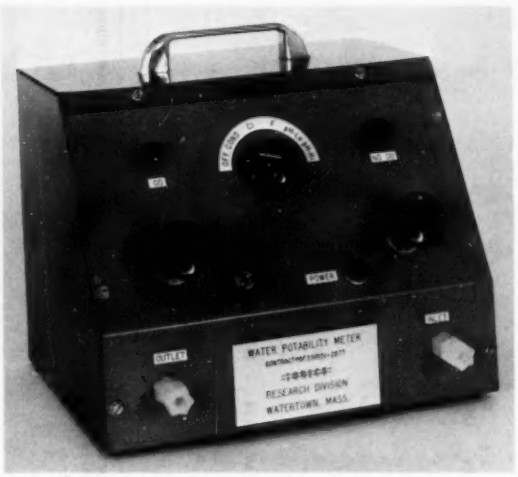


Fig. 9 Potability meter manufactured by Ionics, Inc., Watertown, Mass.

Water-Recovery Function Design Requirements

The design requirements included:

Yield	90% of the H_2O in 15 lb of urine per day on daily basis for 30 days
Gravity	Normal earth and zero gravity
Temperature	15.6 to 35°C
Pressure	380 to 760 mm Hg
Relative humidity	30 to 80%
Atmosphere	Normal earth and 100% oxygen
Acceleration	15 cps at 8-gravity environment in any plane
Water storage	5 lb of hot water 10 lb of cold water
Waste storage	5 lb of waste liquid 5 lb of urine solids

Conclusions Concerning the Integrated Waste-Management System

1. The vacuum distillation–vapor pyrolysis water-recovery process produces potable and sterile water from urine. Water-recovery efficiencies were very good and averaged 93% for the two longest runs. Of the 15 water samples recovered by the vacuum distillation–vapor pyrolysis process (Table 1), three were obtained from units that were electrically heated, seven from units that were radioisotopically heated, and five from units that were heated by both electric heaters and waste heat. Eight of the samples were contaminated either by the failure of the evaporator liquid-level sensor or by improper cleaning after failure. The results of the remaining seven good samples were used as evidence of the workability of the system. Several tests were successfully

conducted using simulated urine (5000 ppm NH_3 in water).

2. The use of radioisotopic heat sources in space-vehicle vacuum distillation–vapor pyrolysis water-recovery systems has been demonstrated to be preferable to other heater types (electric or liquid).

3. Liquid-level controls presented the most difficult problem in the water-recovery system. Simplified methods with minimal electronics proved to be the best approach.

4. Urine solids are extremely abrasive and require special handling procedures. The solids are detrimental to seals, bearings, and close-tolerance devices.

5. Static phase separators in the urine evaporator are preferable to dynamic devices because of power considerations and the abrasive characteristics of the urine solids on moving parts.

6. Feces storage by the slinger method and vacuum drying proved to be an acceptable and sanitary method for space-vehicle feces waste management. Earth-type collection procedures with no handling of bagged feces proved psychologically advantageous.

7. The potability meter is of great psychological benefit to the crew, who must drink water recovered from urine.

Recommendations for the Total Waste-Management System

1. Further evaluation is required to determine the optimum catalyst for the pyrolysis process in the water-recovery system. Catalysts such as platinum, ruthenium, and rhodium have been used successfully. Better catalysts may lower the required operating temperature, but lower temperatures may not be sufficient to sterilize the vapor. Both reducing and oxidizing atmospheres have been used in the pyrolysis chamber. Exact benefits of each are not known, and further study is required.

2. More activity in the area of controls, especially liquid-level sensors for zero-gravity environments, is required.

3. More evaluation of static and dynamic phase separators in a zero-gravity environment is required.

4. The system described herein is sufficiently developed to show its desirability as a flight system. It is recommended that a flight experiment of this type be incorporated in space flights within the near future so that the unit will be space proven before it is required for long-duration space missions. (JEC)

RADIOISOTOPIC COMBINED MOISTURE–DENSITY METER

A radioisotope gage for simultaneous determination of moisture and density in soil has been developed in Hungary by Á. Z. Nagy and T. Rázga [*Journal of Scientific Instruments*, 43: 383-387 (1966)]. A single 8-curie Ra–Be neutron and gamma source is located in the nose of a probe, although neutron–gamma source combinations such as ^{241}Am–Be with ^{137}Cs might be used. In the 45-mm-diameter tube, behind the source, is a lead shield to reduce background, followed by the neutron detector, the gamma detector, a common multiplier, and a pulse-shape discriminator. The neutron detector is a ZnS–B–polyester scintillator, and the gamma detector is a transparent plastic scintillator. These are coupled to the multiplier and produce signals of different shapes at the multiplier output, one shape being proportional to density and the other to moisture content. In tests made with basic building materials to which known amounts of water were added, the results showed a standard deviation of ±1% for moisture contents of 0.02 to 0.9 g/cm³ and densities of 0.8 to 2.5 g/cm³ with counting times of 10 and 1 min, respectively. However, the accuracy was only 4 to 5%, and a better calibration method is needed. (MG)

SOLUBILITY MEASUREMENT WITH RADIOISOTOPE INSTRUMENT

In a method described by Polish scientists, the solubility of cadmium in molten cadmium chloride was determined* by following the decrease in transmission of gamma radiation through the solution with increased solute concentration. The apparatus contains a ^{170}Tm source, whose collimated gamma radiation passes through the solution under study to a scintillation counter. As cadmium metal was added to the melt, the plot of a function of the transmitted radiation count rate vs. concentration was linear until the solution became saturated; at this point there was a definite discontinuity. Solubility values at temperatures in the range 550 to 850°C agreed well with other published values. (MG)

*J. Moscinski and L. Suski, Radiometric Determination of the Solubility of Cadmium in Molten Cadmium Chloride, *Journal of Physical Chemistry*, 40: 1727-1731 (1966).

Section

VI

Isotopes and Radiation Technology

Applications in Life Sciences

Current Status of Nuclear Medicine in Japan

By Masahiro Iio, M.D.*

In Japan, which has the world's second largest nuclear medicine association (1200 members), the use of isotopes in medicine is continually increasing. Since its organization in 1961 with 400 members, the Japanese Society of Nuclear Medicine has held seven annual meetings with a continuously increasing number of abstract contributions:

1961	99	Tokyo
1962	62	Tokyo
1963	111	Tokyo
1964	114	Tokyo
1965	134	Kyoto
1966	148	Tokyo
1967	149	Tokyo

In 1966, of the 52 university hospitals, 50 (96%) were using radioisotopes for both diagnosis and therapy. This is an increase from 78% in 1964 and 90% in 1965. More than 82% of the members of the Society are on the staffs of the university hospitals. At present the members are 34% physician, 19% surgeon, 18% radiologist, 5% gynecologist, and 24% others. There are divisions of nuclear medicine in many universities. The Society has an annual meeting and publishes quarterly the *Japanese Journal of Nuclear Medicine* and one full English edition of *Proceedings of Japanese Nuclear Medicine*.

The first Radioisotope Conference in Japan was sponsored by the Japan Atomic Industry Forum in 1955, the year that radioisotopes were introduced into Japan. Since then this Forum has held conferences every year and a half. A section on medicine and pharmaceuticals is included in the Forum, and medical papers were presented there before the Society of Nuclear Medicine was organized. Since the organization of the latter in 1961, supported by the Japan Radioisotope Association for the first 3 years, these two organizations have cooperated closely. Many foreign visitors attend the meetings, some 12 visitors being expected in the field of nuclear medicine in 1967 from the United States and from the southeast Asian countries.

Radioisotope Techniques in Medicine

Even though the research on sophisticated methods using radioisotopes is carried out mainly at the leading universities, routine radioisotope tests are performed by numerous hospitals. In 1966 some 300 hospitals (including the 50 university hospitals) were using radioisotopes for diagnosis and therapy. Sixty per cent of the 89 nonuniversity hospitals with more than 500 beds, 36% of the 230 hospitals with 300 to 500 beds, 11% of the 326 hospitals with 200 to 300 beds, and 4% of the 770 hospitals with 100 to 200 beds are currently using radioisotopes routinely.

Scanning services are performed in 180 of the hospitals with more than 300 beds. The introduction of simple in vitro tests and small instruments, such as the T-3† and the T-3 meter, was one of the major factors for the rapid increase in the number of hospitals using radioisotopes. Even in the 3400 small hospitals, with 20 to 100 beds, the trend is in the direction of greater isotope use although only 1.1% of these are now using radioisotopes.

The increase in use of radioisotopes over the past 3 years was observed not only in the main island of Japan but also in Hokkaido, Shikoku, and Kyushu, where the number of hospitals using radioisotopes increased threefold.

*Chief of Division of Nuclear Medicine, 2nd Department of Medicine, University of Tokyo. Dr. Iio was at the Johns Hopkins Hospital, Division of Nuclear Medicine and Department of Medicine, Baltimore, from 1961 to 1963. He is now Secretary of the Japanese Society of Nuclear Medicine.

†T-3 stands for Triiodothyronin, used for thyroid-function tests.

Import and Production of Radioisotopes and Radiopharmaceuticals

The demand for radioisotopes has increased continually. Domestic production started in 1963, but this still meets only a minor portion of the demand, 80% of which is used for medical purposes. In 1952, 10 million yen ($28 thousand) was spent for radioisotope imports, more than 50 million yen ($140 thousand) in 1956, and more than 100 million yen ($280 thousand) in 1957. In 1965, 235,000 curies of radioisotopes were imported from abroad: 39% from the United States, 31% from the United Kingdom, 22% from Canada, 1.6% from France, and the rest from other countries. Short-lived radionuclides are produced by domestic reactors to fill special requests by physicians. The Japan Atomic Energy Commission, in its second 10-year plan, is planning to increase the domestic production of radioisotopes through the Radioisotope Center, organized in 1965 at the Japan Atomic Research Institute at Tokai, where four reactors are already in operation.

In the past, 15 purified nuclides have been produced and supplied on demand—^{24}Na, ^{32}P, ^{35}S, ^{42}K, ^{56}Mn, ^{64}Cu, ^{72}Ga, ^{76}As, ^{82}Br, ^{131}I, ^{133}Cs, ^{140}La, ^{153}Sm, ^{177}Lu, and ^{198}Au. In the new plan the following radioisotopes will be produced in sufficient amount to cover the nation's entire demand within 5 years: short-lived ^{24}Na, ^{42}K, ^{64}Cu, and ^{82}Br; medium-lived ^{86}Rb and ^{203}Hg; longer lived ^{65}Zn, ^{110}Ag, and ^{60}Co; and isotopes in high demand, i.e., ^{32}P, ^{131}I, ^{198}Au, ^{35}S, and ^{14}C. Separation of fission products is also planned in the next 5 years.

Currently, raw materials for radioisotope production are predominantly supplied by import. Further processing to radiopharmaceuticals, radiochemicals, and labeled compounds is carried on by two domestic companies. Some products are exported to the southeast Asian countries.

In Table 1 are shown the amounts of radioisotopes used in 1964–1966. The amounts of ^{131}I, ^{32}P, ^{51}Cr, ^{35}S, ^{59}Fe, ^{198}Au, ^{24}Na, and ^{42}K show neither significant increase nor decrease, indicating that requirements for these isotopes had reached a plateau in 1964. The ^{131}I-labeled pharmaceuticals show a similar tendency with the exception of ^{131}I-labeled macroaggregated albumin, consumption of which has increased more than 300% in the past 3 years. However, the use of ^{3}H, ^{133}Xe, ^{75}Se, ^{85}Kr, ^{203}Hg, ^{64}Cu, ^{99}Mo, ^{99m}Tc, ^{125}I, and ^{45}Ca increased significantly[1]—from 200 to 1000%. T-3 kits showed the most significant increase in use during this period, 12,000 kits being used in Japan in 1966.

Table 1 RADIOISOTOPE USE IN JAPAN IN 1964–1966

Radioisotope	Amount used, mc		
	1964	1965	1966
^{32}P	20,950	19,990	19,380
^{131}I (sodium iodide)	54,300	61,850	64,210
(labeled compounds)	4,430	5,300	6,380
^{198}Au	34,940	36,100	47,460
^{51}Cr	920	1,000	860
^{45}Ca ($CaCl_2$)	170	230	140
^{3}H	51,000	82,000	356,000
^{125}I	190	270	360
^{133}Xe	1,670	2,360	2,600
^{59}Fe	390	240	170
^{42}K	260	300	360
^{64}Cu	89	83	184
^{85}Kr		1,910	8,100
^{24}Na (NaCl)	410	500	410
^{203}Hg (chlormerodrin)	333	480	490
^{75}Se		37	146
^{131}I T-3 kits	3,005	6,772	11,963
^{125}I Immuno assay kits		19	94
Generator (^{99m}Tc)		2,150	8,050
^{35}S	1,350	1,030	1,280

Instrumentation

Because of its world-famous basic electronics industry, Japan is in a favorable position to supply all types of instrumentation. Half a dozen industries are oriented for the production of instruments for nuclear medicine. Various types of scintillation scanners with 3- or 5-in.-diameter crystals are manufactured by three companies with a variety of accessories, and about 180 scanners for routine use are available. The use of scanning is increasing rapidly in Japan, and scanners have penetrated city hospitals and private hospitals as well as the university hospitals. Handy equipment such as the T-3 meter is among the best-selling equipment in Japan. A liquid scintillation spectrometer is manufactured by four factories and is contributing to the increase in use of this technique in nuclear medicine.

"Whole-body" counters are installed in three institutes, and a good 800-channel analyzer is available commercially. Besides a variety of scalers and analyzers for nuclear medicine, unique instruments have been devised and manufactured and are now sold commercially. For example, the author's group[2] has produced a catheter-type semiconductor detector 3 mm in diameter and 1–1.5 m long with a low-noise amplifier system for use in hemodynamic studies and in detection of gastrointestinal malignancy.

Many other instruments have been reported, e.g., the medical Universal Human Counter,[3] new gamma cameras,[4] and color scanners.[5] The application of computers is becoming an important part of nuclear medicine, with the aid of six domestic manufacturers.

International Relations

Because of the increasing necessity of international exchange of information in this field, the Japanese Society of Nuclear Medicine sends two persons every year to the Annual Convention of the Society of Nuclear Medicine in the United States. In addition, more than 10 members of the Japanese Society of Nuclear Medicine have attended the Convention each year for the past 3 years, presenting papers and then visiting various institutes in the United States.

In accordance with the movement to organize the International Federation of Nuclear Medicine, Japan is organizing an Asian Federation as a part of the International Federation. We have been gratified and impressed by the strong support for organizing such federations regionally in Asia. Of the six countries now interested in such a liaison, communication with the Korean Society of Nuclear Medicine is one of the closest. There is continual exchange of doctors between the two societies.

• • •

Japan has an increasingly active field of nuclear medicine. There is still a shortage of facilities and funds, but enthusiastic young researchers are overcoming these difficulties. It is our hope and plan that this field will be supported as a science project of our government and that the worldwide exchange of knowledge, personnel, radioisotopes, and instrumentation will increase.

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Section

VII

Isotopes and Radiation Technology

Isotopes Around the World

Radiation and Radioisotopes in Canada*

Radiation technology in Canada in 1967 may be just on the inflection point of a growth curve typical of many new industries, believes Roy Errington, vice-president of Commercial Products, a division of Atomic Energy of Canada Limited (AECL): "A long period of 15 years at a relatively small dollar volume, followed by many years at a high dollar volume." Mr. Errington's two presentations to the 11th AECL Symposium on Atomic Power, one on history and current scale of the Canadian radioisotope program and one on its future perspectives, gives the background of his organization's renewed drive to promote more rapid progress in exploitation of radiation techniques in industry, science, and medicine in Canada.

Commercial Products, the chief organization in Canada for production and distribution of radioisotopes, enjoyed a record year in 1966, with total shipments of about a million curies, averaging 14 individual orders per day by the close of the year and totaling 6 million curies since shipments began in 1953.

Installation of a medical-supplies-sterilization irradiator for New Zealand in late 1966 brought the Commercial Products total of industrial-scale installations of this type to seven. Two pilot-scale food irradiators were shipped to India, joining 170 AECL research irradiators at work in 30 countries.

The main business of Commercial Products remains in the field of ^{60}Co cancer therapy (Fig. 1). There are now over 600 units in 47 countries, and medical-grade high-intensity cobalt sources make up the largest single export item. And, as in the past few years, exports continue to make up 90% of Commercial Products' business (Fig. 2). As long as this situation prevails, it is unlikely that private enterprise will be attracted into the business of producing and distributing radioisotopes. According to Errington, "Most Canadian industries expect to live on domestic sales and take plus values from export sales. There is not enough Canadian market to make this practical."

However, the areas now opening up in the applications of radiation in industry, science, and medicine already provide a substantial near-term challenge to progressive private industry in Canada. The curve showing anticipated growth of nuclear instrumentation in the United States reveals an almost asymptotic rise anticipated over the next 2 or 3 years. Prices of isotopes and equipment to utilize them are decreasing

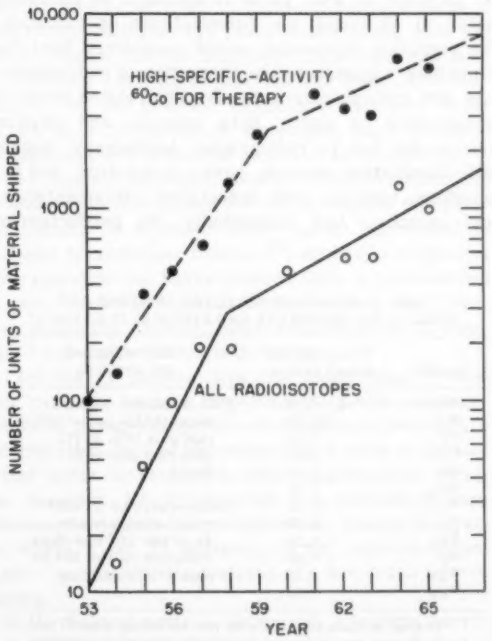

Fig. 1 Growth rate of AECL radioisotope shipments.

*Reprinted, with minor editorial changes, by permission of the editors from *Canadian Nuclear Technology*, 6(1): 14-17 (January–February 1967).

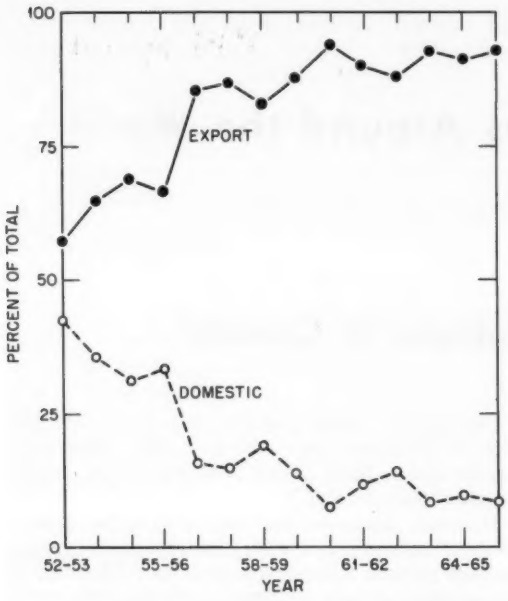

Fig. 2 Relative value of revenue from Canadian export and domestic radioisotope sales.

(Table 1). When a ^{60}Co gamma source can be had for as little as $60, price is no longer an inhibiting factor in exploring the potentialities of radiation. The radiation processing areas considered best for immediate exploration are production of radiochemicals and radiopharmaceuticals. The related areas of manufacture of alpha, beta, gamma, and neutron sources for use in radiography, teletherapy, gages, self-illuminating devices, power generation, and irradiators, together with associated instrumentation, hold promise. And, undoubtedly, the production of reactors, cyclotrons, and fission-product radioisotopes will increase.

Already at least one U. S. firm is promoting industrial radiation processing using electron accelerators. Opinion on the economics of using such equipment in lieu of radioisotopes is divided, and the key to the choice in any individual case seems to be the surface-to-volume ratio of the material to be irradiated and the dosage required. Experts are now canvassing Canadian industry for radiation applications.

According to Commercial Products there is no shortage in Canada of any grade or variety of radioisotope. "Production in NRX and NRU for all radioisotopes is sufficient to supply Commercial Products' present and short-term requirements," says C. H. Hetherington, Director of Marketing. "For the future, power reactors, multipurpose reactors, and isotope production reactors will likely be considered and used as the market requirement develops." The main task ahead, then, is to stimulate greater domestic application of the important national resource of nuclear radiation. Commercial Products is initiating a number of cooperative research and development programs to meet the regional needs of all parts of Canada. More emphasis will be placed on basic research and on the increasing participation of industry, federal and provincial government departments, and, especially, universities. These efforts will be backed up not only by the new AECL mobile radiation laboratory and already familiar mobile irradiator but also by a number of pilot-scale irradiation plants. The first full-scale commercial food-irradiation plant, previously operated by Newfield Products, near Montreal, will also be operated by Commercial Products to do pilot and custom irradiations for industrial and research organizations.

Besides the isotopes mentioned in Table 1, Commercial Products also sells, on a large scale,

^{198}Au	^{192}Ir	^{197}Hg
^{131}I	^{55}Fe	^{203}Hg

and on a smaller scale,

^{122}Sb	^{64}Cu	^{191}Os	^{111}Ag
^{124}Sb	^{152}Eu	^{103}Pd	^{85}Sr
^{125}Sb	^{154}Eu	^{210}Po	^{182}Ta
^{131}Ba	^{155}Eu	^{143}Pr	^{97}Tc
^{210}Bi	^{71}Ge	^{147}Pm	^{127}Te
^{115}Cd	^{199}Au	^{149}Pm	^{204}Tl
^{131}Cs	^{181}Hf	^{186}Re	^{170}Tm
^{137}Cs	^{114}In	^{86}Rb	^{113}Sn
^{45}Ca	^{197}Hg	^{97}Ru	^{185}W
^{141}Ce	^{99}Mo	^{103}Ru	^{90}Y
^{36}Cl	^{147}Nd	^{46}Sc	^{65}Zn
^{51}Cr	^{59}Ni	^{75}Se	^{95}Zr
^{58}Co	^{63}Ni	^{110}Ag	

Table 1 PRICES OF SELECTED ISOTOPES AND ASSOCIATED EQUIPMENT AND SERVICES IN CANADA*

Isotope	Price† per mc (small orders)	Other equipment and services
^{134}Cs	$ 1.35	^{60}Co irradiator (Gammabeam 150A): output 500 rhm plus 10%, $8475; 1500 rhm plus 10%, $9135
^{36}Cl	350.00	
^{14}C	7.00	
^{51}Cr	2.15	
^{125}I	3.25	
^{60}Co	0.55	
^{59}Fe	27.00	Neutron service irradiations: small capsule, $5.40 per 10^{13} flux-days, minimum charge $54.00 plus $27.00 handling
^{32}P	0.86	
^{85}Sr	16.20	
^{35}S	0.65	
^{65}Zn	2.15	
^{45}Ca	7.00	

*Prices in Canadian dollars, not including federal tax, f.o.b. Ottawa, and including disposable shipping containers.
†Plus handling charge of $20 per unit (vial or other container).

Applications in Industry

Industrial irradiation in Canada seems to have come of age, and the fact that unforeseen imbalances in the potato market over the past 2 years caused the demise of Newfield Products does not invalidate the technical success and market acceptance of potato irradiation. The Ethicon operation at Peterborough for sterilization of medical supplies by radiation seems to be eminently successful.

Food, according to Dr. A. B. Lillie, head of Commercial Products' Science Branch, shows the most potential for applications of large radiation sources. Its total sales value in this country is about 15% of that of all manufacturing. Our short growing season, long shipping distances, and substantial export trade make especially valuable any techniques that will extend product shelf life. Now under investigation in Canada (besides potatoes and onions, which have already been approved for irradiation processing by the Food and Drug Directorate) are sugar beets, cereals, fruits, fish, poultry products, meat, and mushrooms.

While the major radiation effect utilized in present experimental applications has been biocidal—destruction or inhibition of growth of sprouts, plants, or animal parasites—work funded by Commercial Products is also being pursued at the University of Manitoba on a Russian-discovered effect, i.e., that low-level irradiation of seed increases grain crop yields. There is considerable Canadian interest in a similar new technique being explored in the United States—irradiation of seed stock to stimulate growth of potato tubers, possibly permitting the production of two crops annually.

Although there are apparently successful applications of radiation processing in the chemical industry in the United States (most notably in production of ethyl bromide at Dow Chemical's Midland, Mich., plant), there are no similar applications in the Canadian chemical industry as yet. Commercial Products is, however, sponsoring a research project at Queen's University on radiolysis of hydrocarbons, and a program on radiation-catalyzed chlorination of hydrocarbons is being carried on at Waterloo University.

Extensive work has been done in the United States on radiation grafting of polymers to wood, but actual marketing of the product is in too early a stage to assess its commercial acceptance. Work is going on in Canada in this area, but Dr. Lillie of Commercial Products states, "We are currently slowed down by our temporary inability to provide large enough samples for industrial evaluation. This should be alleviated by the pilot-scale irradiator program."

Pioneering work was conducted some years ago at the University of Toronto by a team under Dr. Howard Rapson on grafting of styrene and other monomers with textile fabrics. Research with a variety of monomers and substrates is being carried on, but there has not yet been any commercial application. Recent work on radiation-grafted nylon fabrics and radiation-resistant greases has been described by Dr. Sharda DasGupta.[1]

Applications in Research

At the 11th AECL Symposium on Atomic Power, R. W. Tolmie indicated world usage and annual growth rates for the four basic categories into which most uses of nuclear radiations for scientific instrumentation fall:

	Millions of dollars (1966)	Annual growth, %
Tracing	408	33
Gaging	336	15
Radiography	126	25
Element analysis	54	52

The mean annual growth rate for all applications, weighted by category size, is 26.5%.

In industrial application of tracer techniques, an interesting experiment was carried out by the Pulp & Paper Research Institute of Canada. Instead of mixing a radioactive tracer isotope with the material under study, the material itself—a section of pipeline—was activated by a neutron flux. The pipe was part of a pilot-plant installation to study long-distance transport of wood chips by pipeline. After the activated section was reinstalled, a thousand-gallon sample of water-chip mixture was taken downstream from the active section, and the activity of the sample was used to calculate the rate of pipe-surface wear. However, still by far the greatest application of tracer techniques is in laboratories, especially those in which biochemical and biological research is carried out.

For gaging purposes, Commercial Products has developed an instrument for measuring the mass per unit length of synthetic fiber yarn. The thread is exposed to radiation from a ^{125}I source, and some of this radiation is backscattered into a pressurized krypton ionization chamber surrounding the fiber. The ionization current, after amplification, is measured to indicate the yarn thickness.

Low-energy gamma rays like those from ^{125}I also hold promise for use in plating-thickness measurements or in two-component elemental analysis, where the radiation excites characteristic X rays in the material under investigation. Some applications, using, for example, Hf–Zr sources in a portable X-ray-fluorescence analyzer configuration, include analysis to within 1 or 2% accuracy of tin, zinc, or copper plated on steel to thicknesses of a few dozen microinches.

In the field of neutron activation, a neutron source using ^{124}Sb–Be sources develops neutron fluxes of 10^8 n/(cm^2)(sec). This convenient source of neutrons

should help broaden the applications of neutron activation techniques, since neither an expensive neutron generator nor a long trip to a research reactor is required.

Dr. K. W. Geiger of the National Research Council's (NRC) Division of Applied Physics has contributed to nuclear instrumentation technology a neutron dosimeter of a much lighter weight than that heretofore used. The instrument incorporates a cadmium shell 4 in. in diameter inside a polyethylene sphere less than 8 in. in diameter. Previous dosimeters used a 12-in.-diameter plastic sphere, weighing three times as much as the new model.

One result of the general scarcity of Canadian literature on radiation applications in this area is that no comprehensive source exists from which any indication could be gained as to what use is being made of the technology in science and industry as compared to potential use. Dr. Tolmie estimates that, right now, radioisotope applications are saving Canadian research and industry "between $10 million and $20 million a year."

Applications in Medicine

The application of ^{60}Co in cancer therapy is well established, being the oldest as well as the single largest volume application of reactor-produced isotopes. As noted above, some 600 teletherapy machines have been sold by Commercial Products in the past decade, and new installations are made every month, although the rate of growth is now flattening slightly.

Other medical applications of irradiation being investigated include the use of gamma radiation for making influenza vaccine. Polley[2] (Virus Laboratories, Department of National Health and Welfare) has found that irradiation inactivates influenza and mumps virus without destroying the immunity-giving hemagglutinin fraction. Thus vaccines can be prepared quickly and with precise control of the inactivation.

In the field of microbiology, spores of foulbrood and other bee diseases in honey and honeycomb as well as yeasts causing fermentation were effectively controlled by ^{60}Co irradiation.[3]

Research in radiobiochemistry included investigations by Vaisey and Thatcher[4] on the mechanism of radiation attack on adenosine triphosphate (ATP), an important constituent of animal protein. Other work done at the Food and Drug Directorate's Ottawa laboratories included a study of the effect of gamma radiation on the sedative pentobarbital.[5]

Note Added in Proof: A later report [*Canadian Nuclear Technology*, 6(5): 35 (September–October 1967)] states that of a record revenue of $8,300,000 of the AECL's Commercial Products group, some $7,500,000 came from exports. Commercial Products' 45 agents covering 130 countries realized much of its business from shipments of more than 1 million curies of radioactive material, chiefly ^{60}Co for cancer therapy units but with an increasing amount of this cobalt going to industrial radiation applications. During the year AECL completed and installed a plant in New Zealand for sterilization of medical supplies and shipped two smaller irradiators to India. (MG)

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Production and Use of Radioisotopes in Spain, 1964-1965*

The value of radionuclides distributed in Spain reached 7.5 million pesetas ($125,000) in the 2-year period 1964–1965, compared to 4 million pesetas ($67,000) in 1962–1963. The increase in curies produced and number of deliveries made since 1957 is shown in Fig. 1. This does not include imports, which were chiefly the ^{60}Co for the Nayade irradiation facility (4320 curies) and for teletherapy sources. In 1965 the number of persons authorized to use radioisotopes was 299 and the number of installations, 194.

*Based on Biennial Report 1964–1965, Junta de Energia Nuclear, Ministerio de Industria, Spain, pp. 153-167 (in Spanish). Mr. F. de la Cruz Castillo, Head of the Nuclear Chemistry Division, Chemistry and Isotopes Department, kindly reviewed this summary before publication.

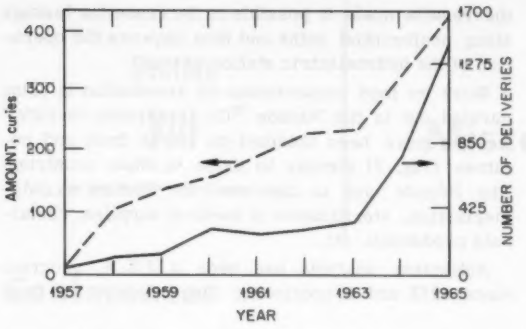

Fig. 1 Distribution of radioisotopes in Spain, 1957–1965.

Production

RADIONUCLIDES

Isotope production in Spain was started in 1959 in the JEN-1 reactor. In the period 1962–1965 the amounts of some important isotopes produced increased considerably (Table 1), for example, ^{32}P 3-fold, ^{60}Co 100-fold, and ^{82}Br 10-fold. In addition, as both the reactor power increased and downtime decreased, products of higher specific activity were produced, and additional nuclides were added to the program. For example, the activity concentration of colloidal ^{198}Au was increased from 20 to >40 mc/ml, making it possible to ship the product long distances and still retain usable activities. The large increase in ^{82}Br production was in response to the demand for this isotope for hydrological research, important in Spain's dry climate.

Table 1 PRODUCTION OF RADIOISOTOPES IN SPAIN 1962–1965

Radioisotope	Amount, mc			
	1962	1963	1964	1965
^{124}Sb		1,092	15,592	4,175
^{35}S	646	259	592	252
^{131}Ba		0.1	0.05	
^{82}Br	350			33,512
^{14}C			13.1	
^{36}Cl			0.02	
^{58}Co	1.2		4.5	
^{60}Co	1		12.6	106.2
^{64}Cu			185	
^{51}Cr	377	754	421	329
^{32}P	5,200	4,850	8,711	15,310
^{59}Fe	0.1	0.4	1.9	49
^{192}Ir (source)	8,178	2,907	5,931	16,529
^{193}Ir (wire)	76	66	382	338
^{90}Y	7		31.2	
^{95}Zr		0.02		
^{140}La	32			2.6
^{54}Mn	0.1	0.4	5.6	7.2
^{197}Hg				1,272
^{203}Hg	69	40		
^{198}Au (colloidal)	45,970	40,503	71,746	79,452
^{198}Au (granular)	5,548	3,309	2,397	4,193
^{198}Au (wire)	437	119		
^{86}Rb	19	0.7		
^{103}Ru		0.9	1.4	
^{75}Se				91.5
^{24}Na	3	6		13
^{204}Tl			1	
^{182}Ta			91.5	
^{3}H				3,043
^{131}I	1,769	17,414	29,141	38,375

Better production methods have also been developed. For example, the ^{35}S formed by irradiation of KCl is converted to the sulfate and reduced directly, without prior separation of the KCl. In a new method of producing ^{131}I, the target is tellurium oxide, which is thermally and radiolytically more stable than the telluric acid previously used. Production of ^{131}I in gelatin capsules to facilitate its use in diagnosis has also been started. The specific activity of ^{192}Ir sources for use in gamma radiography has been increased considerably, and 4- by 4-mm sources are now made which have an activity of 25 curies.

A preliminary study has been made on preparation of carrier-free radioisotopes with fast reactor neutrons, and methods are being developed for producing a number of radionuclides, e.g., ^{46}Sc from titanium, ^{54}Mn from iron, ^{58}Co from nickel, ^{59}Fe from cobalt, ^{63}Ni from copper, and ^{64}Cu from zinc. Likewise, methods are being developed for making targets of higher purity than used heretofore, as well as methods for determining the fast-neutron flux at the JEN-1 irradiation positions and the effective cross sections for the nuclear reactions of interest.

In the fission-product field, work has been limited to developing methods for producing ^{103}Ru and for recovering ^{90}Y, completely free of ^{90}Sr, for medical use.

LABELED MATERIALS

Both inorganic and organic labeled materials are made. Among the inorganic materials are ^{32}P-labeled $PSCl_3$ and P_2S_5. The inorganic ^{32}P radiopharmaceuticals include colloidal chromic phosphate and zirconium phosphate and ^{90}Y-labeled yttrium silicate. The ^{35}S-labeled materials include elemental sulfur and labeled cadmium sulfide, H_2S, $PSCl_3$, and anhydrous Na_2S.

The organics are mainly of two types — insecticides and pharmaceuticals. Insecticides labeled with ^{35}S and ^{32}P have been synthesized, special attention being given to both chemical and radiochemical purity. In some cases the labeled product has a considerably higher chemical purity than the commercial, nonlabeled product. The newer radiopharmaceuticals made are ^{131}I-lipiodol and ^{131}I-labeled growth hormone. On a laboratory scale, ^{131}I-thyroxine, ^{131}I-triiodotyrosine, and ^{75}S-selenomethionine have been prepared and will soon be commercially available.

There is a complete installation for making ^{3}H-labeled organic compounds, starting with tritiated water or tritium gas, by synthesis, isotope exchange, or the Wilzbach method. A method has been devel-

oped for labeling thymidine in the methyl group with ^{3}H to give a product of very high specific activity. Labeled thymine is made by a chemical method and converted to thymidine by an enzymatic process.

Uses

INDUSTRY

For industrial application a method has been developed for controlling the thickness of electrolytic deposits of Ni–Sn alloy on iron, copper, or brass. The layer is 1 to 8 μ thick, and when the coated sample is exposed to a bremsstrahlung source, the absorption by the coating of X rays excited in the support is a function of the coating thickness. The measurement takes about 5 min; the error is <5%.

In the field of hydrology, a method for routinely measuring large flows of water in rivers is being investigated. Numerous experiments have been made in rivers to compare values obtained with radioactive tracers with those obtained by conventional methods. In a series of experiments carried out over a year's time on detection of leaks in a reservoir, the results made it possible to decrease the leakage along preferential paths and thus improve the operation of the hydroelectric station served.

Work on food preservation by irradiation is being carried out in the Náyade ^{60}Co irradiation facility.* Results have been obtained on citrus fruit and potatoes (Fig. 2) similar to those in other countries. The Náyade unit is also used for studies on polymerization, sterilization of medical supplies, chemicals production, etc.

Activation analysis has been a J.E.N. program since 1963 and is continuing. The technique has been used for determining Cr, Mn, Co, and Cu in a program on analytical methods; for Nd in ^{147}Pm solutions; for Na, Cl, K, Mn, Cu, and Br in rice from different places; for Br in water; and for Th in uranium ores and in process by-products. It has also been used to identify automobile paint in forensic studies.

(Martha Gerrard)

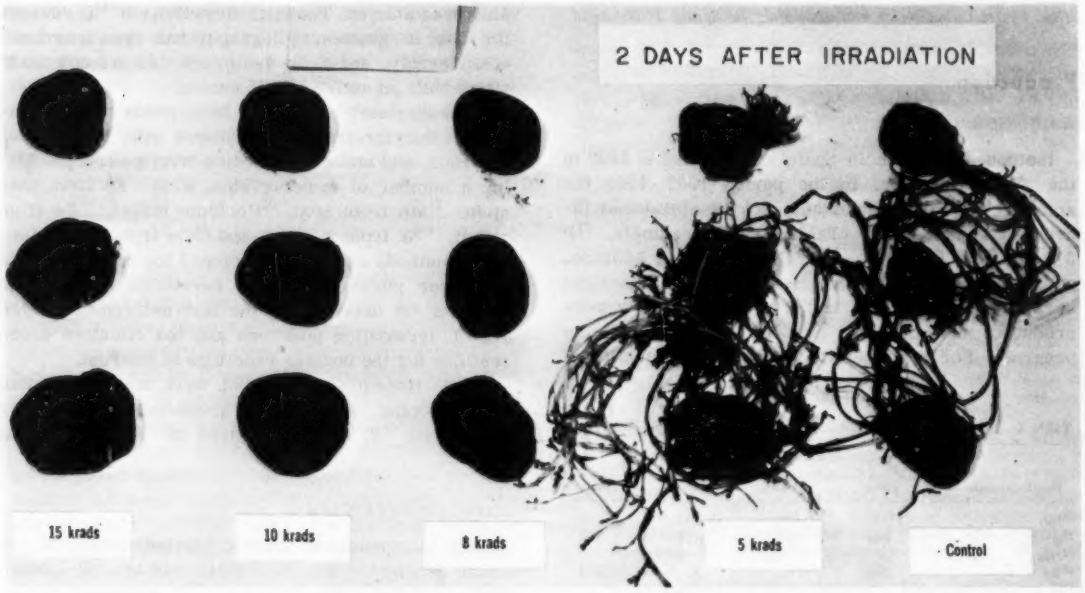

Fig. 2 Gamma-irradiated (^{60}Co) and control potatoes (Gineke) from the Spanish studies.

*Spanish Náyade Irradiation Facility, *Isotopes and Radiation Technology*, 4(4): 428–429 (Summer 1967).

Section VIII

Isotopes and Radiation Technology

Miscellaneous

Conferences

Second International Conference on Medical Physics, August 1969

The Second International Conference on Medical Physics will be held in Boston on Aug. 10–14, 1969, under the sponsorship of the U. S. National Committee for Medical Physics and the International Organization for Medical Physics. The conference will include symposia and papers on radiology and nuclear medicine. Preliminary programs will be available in June 1968. Further details are available from Dr. Edward W. Webster, Secretary-General, Department of Radiology, Massachusetts General Hospital, Boston, Mass. 02114.

AEC Activities

New Shipping Regulations To Ease Radioisotope Transportation

New regulations of the Department of Transportation—still being drafted—will ease restrictions on shipment of radioactive materials and thereby reduce shipping costs by 10 to 20%. The main change will transfer enforcement authority from the AEC to the Department of Transportation for certain quantities of radioactive material.

The limits on shipments under Department of Transportation rules are set at 15 g of radioactive fissionable materials, 5000 curies (sealed), and 200 curies (unsealed). Above these limits, regulations of the AEC will still be applicable. The prior approval of shipments of 20 curies or less has also been eliminated. In addition, the red, white, and blue radioactivity labels will be changed to the UN yellow and white labels. The change most likely to reduce costs is that higher radiation levels will be permitted inside trucks as long as the radiation level outside does not exceed the permitted value.

Tulane University Awarded Brain-Scan Contract

Tulane University has been awarded a $9800 research contract from the AEC to test the use of newer, short-lived radioisotopes for brain scanning. The contract covers research through Aug. 31, 1968. Dr. J. U. Hidalgo will direct the program, which will develop methods to establish the reliability of ^{99m}Tc and other short-lived radioisotopes in brain-scanning technology.

University of Miami Receives Contract To Study Radiopharmaceuticals

The AEC has announced that the University of Miami has been given a contract to examine the effects of new radiopharmaceuticals on various body tissues. The program will be directed by Dr. E. M. Smith, and it is set up to run through Oct. 14, 1968. The specific objective of the program is to study radioactivity distribution in the body after receiving selected diagnostic radioisotopes.

Decrease in AEC Isotope Sales

The effects of the AEC's policy of withdrawing from sales of radioisotopes as industry builds up its production capability are evident from the data published in *Nucleonics Week* (page 5, Nov. 2, 1967) based on data supplied by the Isotopes Information Center of Oak Ridge National Laboratory. Sales in terms of shipments, curies, and dollars are compared in Table 1.

Table 1 AEC ISOTOPE SALES,* FY 1966 AND FY 1967

	Shipments		Curies		Dollars	
Purchaser	FY 66	FY 67	FY 66	FY 67	FY 66	FY 67
Domestic	6,140	2,994	1,053,183	904,000	2,161,458	1,712,153
Foreign	1,134	998	527,756	516,000	712,400	643,947
Government†	1,610	1,118	3,632,607	2,947,000	2,122,885	1,648,146
Total	8,884	5,110	5,213,546	4,367,000	4,996,743	4,004,246

*Includes cyclotron and reactor service irradiations.
†Includes transfer between AEC facilities.

Fee for Radioisotope Courses Raised

Courses conducted for the AEC by Oak Ridge Associated Universities in radioisotope techniques will cost more for students effective July 1, 1968. Individuals from profit-making organizations will have their fees increased from $100 per week to $150 per week. Government employees and members of

nonprofit organizations will have to pay $75, an increase from $50. For university personnel, fees for the radioisotope course will remain at $25 per course. This is the first increase in fees since they were set in 1962; it reflects increasing costs of the program, mainly stemming from attempts to keep up with current developments in radioisotope applications. The fees do not represent full costs of the courses, which are partially financed by the AEC. Current course material includes use of radioisotopes in research, in special applications, and in medical diagnosis.

Mobile Radioisotope Laboratory Travels Coast to Coast

In 1968, students and faculty of colleges across the United States will be attending concentrated 2-week courses in radioisotope techniques and applications as a result of the Oak Ridge Mobile Radioisotope Laboratory program. During the first 6 months of the year, the three mobile units will visit 30 colleges and universities in Colorado, Florida, Idaho, Kansas, Maryland, New Jersey, New York, Pennsylvania, Utah, and Washington. The program is operated by Oak Ridge Associated Universities for the AEC. Each course presents a 2-week series of laboratory experiments and lectures. Lectures treat radiation detection, instrumentation, nuclear theory, radiological safety, and principles of a wide variety of radioisotope applications. Experiments deal with isotope dilution, radiochemical separation techniques, and biological studies. More than 200 schools in 37 states have requested and received the mobile laboratory course since the program began in 1959. For further information, contact Special Training Division, Oak Ridge Associated Universities, P. O. Box 117, Oak Ridge, Tenn. 37830.

Radioisotope Sales at Oak Ridge Set New Record—Again

For the fourth straight calendar year, the AEC's Oak Ridge National Laboratory has set a record for radioisotope sales. The laboratory sold 3,669,646 curies of activity in 1967, more than a million curies greater than in calendar 1966 and only slightly less than triple the amount sold in calendar 1965. The total number of shipments continues to drop because of AEC's policy of turning over to industry the routine production and distribution of radioisotopes when ability and capacity are demonstrated by industry. In 1967, shipments numbered 3956.

Three More Booklets Join the "Understanding the Atom" Series

The AEC has published three additional booklets in its "Understanding the Atom" series. "Animals in Atomic Research" by E. R. Ricciuti discusses studies of the effects of nuclear radiation on life. Mice, rats, pigs, hamsters, cats, rabbits, and dogs as well as unusual animals ranging from amoebas to alligators have been used extensively. The 52-page booklet describes the types of radiation, radiation and life, genetic effects, radiation in the environment, radiation safety, beneficial applications, veterinary medicine in the nuclear age, germ-free animals, and care of laboratory animals.

"The First Reactor" recounts the events of 1942 concerned with man's achieving the first self-sustaining nuclear reaction. The 43-page booklet includes comments by Enrico Fermi, who led the experimental team, and a personal memoir by Mrs. Fermi. There are also photographs of the CP-1 pile in various stages of construction.

"The Chemistry of Noble Gases" by C. L. Chernick describes the noble gases, traces the early history of attempts to form compounds, describes the preparation of the first xenon compounds and compounds of the other noble gases, and shows how noble gas compounds are used. A copy of this 48-page booklet, or any other booklet in the series, is available free from the U. S. Atomic Energy Commission, P. O. Box 62, Oak Ridge, Tenn. 37830.

AEC Withdraws from Production of Low-Specific-Activity ^{60}Co

The AEC announced its intent to withdraw from production and distribution of ^{60}Co with a specific activity of 45 curies/g or less Jan. 27, 1968. The AEC has determined that there is adequate private capability for production of this grade ^{60}Co to meet domestic needs and to ensure effective competition in marketing. The private companies producing ^{60}Co are General Electric Company and Neutron Products Incorporated; the firms estimate that they produce 3 Mc annually. The policy of the AEC is to withdraw from isotope production whenever private industry demonstrates that it has the capacity to replace AEC production.

As with other withdrawals, AEC will continue to meet requirements when the purchaser certifies a need for material not available commercially.

Colorado Joins List of Agreement States

Effective Feb. 1, 1968, Colorado assumed regulatory responsibility from the AEC for radioactive materials within the state's borders. The agreement was signed in Denver by Governor J. A. Love and AEC Commissioner G. F. Tape on January 16. The transfer of authority will include licensing, rule making, and enforcement of rules covering uses of radioisotopes; the source materials uranium and thorium; and small quantities of fissionable materials. There are about 225 AEC licenses in Colo-

rado for the use of radioactive materials. The State Health Department already has authority over X-ray machines and radium, and the State Bureau of Mines exercises authority over radiation safety in Colorado's uranium mines. Colorado is the 18th state to assume regulatory authority from the AEC.

Food-Irradiation Petitions Withdrawn, Petition Requirements Explained

Two food-irradiation petitions to the Food and Drug Administration (FDA) have been withdrawn. One submitted by the AEC asked for approval of the use of gamma radiation to preserve fresh strawberries, and the other, submitted jointly by the AEC and the Army, asked approval of the use of gamma radiation to preserve lemons and oranges. Specific reasons cited for the withdrawal were lack of 2-year feeding studies for the strawberries and insufficient storage and shipping data for the citrus fruits.

These actions followed several meetings between FDA and AEC officials. The FDA has indicated that it is not making the requirements for approval of irradiated food any stiffer; the FDA is saying that 2-year feeding requirements are absolute. FDA claims that more than 50% of all food-additive petitions have to be refused—whether radiation, chemical, or whatever.

A significant result of these explained requirements is a 2- to 3-year slippage in the commercial availability of irradiated food.

Availability of Isotopes and Services

ORNL Stable-Isotope Inventory Augmented

Recent additions to the ORNL enriched-stable-isotope inventory include the following:

Isotope	Purity, %	Amount available, g
^{33}S	17.5	8.6
	>70	0.2
^{84}Sr	82.2	17.6
^{87}Rb	99.2	34.8
^{98}Mo	98.3	305.1

NBS Announces Revised Prices for Standard Reference Materials

The Standard Reference Materials Program of the National Bureau of Standards (NBS), which dates from 1905, has been placed on a fully self-supporting basis. Before Dec. 15, 1967, costs were partially subsidized; beginning Dec. 29, 1967, the customer will pay the total program costs—including production, certification, and distribution of each standard material. The NBS still furnishes major, long-term support for the program through its appropriated research funds at a cost of about three times the value of the reimbursable efforts put into the program.

Price changes affecting the isotope industry are given below, taken directly from the NBS memorandum of Dec. 22, 1967.

Category 3.22 Isotopic Reference Standards

SRM No.	Kind	Price, dollars
975	Sodium chloride	35.00
976	Copper metal	35.00
977	Sodium bromide	35.00
978	Silver nitrate	35.00
979	Chromium nitrate	35.00
980	Magnesium metal	35.00
981	Natural lead	100.00 per set
982	Equal atom (206/208) lead	
983	Radiogenic lead	

Category 4.5.1 Alpha-Ray Standards

SRM No.	Kind	Price, dollars
4902	Polonium-210	50.00
4904-C	Americium-241	76.00

Category 4.5.3 Beta-Ray, Gamma-Ray, and Electron Capture Solution Standards

SRM No.	Kind	Price, dollars
4921-C	Sodium-22	37.00
4922-E	Sodium-22	37.00
4924	Carbon-14 (water)	43.00
4925	Carbon-14 (benzoic acid in toluene)	43.00
4926	Hydrogen-3 (water)	43.00
4927	Hydrogen-3 (water)	43.00
4929-B	Iron-55	54.00
4940	Promethium-147	55.00
4943	Chlorine-36	38.00
4944-D	Iodine-125	66.00
4947	Hydrogen-3 (tritiated toluene)	41.00
4948	Cerium–praseodymium-144	65.00

Category 4.5.5 Point-Source Gamma-Ray Standards

SRM No.	Kind	Price, dollars
4991-B	Sodium-22	62.00
4997-D	Manganese-54	50.00
4999-D	Cerium-139	55.00
4200	Cesium-137	46.00
4201	Niobium-94	55.00
4203-A	Cobalt-60	65.00
4203-B	Cobalt-60	65.00

Mallinckrodt Contracts To Use University of Missouri Reactor

To be fully competitive with other isotope producers, Mallinckrodt Nuclear has contracted for time in the University of Missouri's new research reactor. The reactor's peak thermal flux of 6×10^{14} n/(cm^2)(sec)

will be used to produce long- and short-lived radioisotopes in two core positions—less than 1% of the radiation capacity. The University announced that Mallinckrodt support will mean that the reactor can now run, and be used by the researchers, 24 hr a day for 5 days a week instead of the previous single shift a day. Although Mallinckrodt's contract is not an exclusive one, the company has expressed interest in financing a 7-day week and using all excess reactor time and irradiation capacity for its isotope-production program.

France Tries for Greater Competition in Radiopharmaceutical Field

To increase its share—presently very small—of the lucrative U. S. isotope market, the French Commissariat à l'Energie Atomique (CEA) will offer American customers a new and varied line of radioisotopes. The agent in the United States will be CAPINTEC (Capital for International Technology). Heading the new line will be a new ceramic cesium therapy seed, and the whole line will be aimed at radiopharmaceutical manufacturers and original equipment manufacturers. CEA will also serve as an outlet for isotopes from Belgium and Italy. Other major exporters to the United States are Canada and England.

General

Guide to Food-Irradiation Services Issued by BDSA

The Business and Defense Services Administration has recently issued a booklet on "Guide to Services, Suppliers, and Research: Food Irradiation." Commercial firms, government facilities, U. S. Army contractors, and AEC contractors are listed. There is a Finding Guide that lists the firms' services. The 35-page booklet is for sale from the U. S. Department of Commerce, Sales and Distribution Branch, Washington, D. C. 20230, for 50 cents.

Teledyne Acquires Con-Rad, Merges It with Isotopes, Inc.

During August 1967, Controls for Radiation, Inc., was acquired by Teledyne, Inc., and subsequently merged with Isotopes, Inc., a Teledyne Company. Isotopes, Inc., will assume the responsibility of providing analytical services to all former Con-Rad customers, and the analytical facilities and personnel of Con-Rad will be consolidated with those of Isotopes, Inc., at its Westwood Laboratories. The analytical laboratory at Westwood is under the direction of Dr. J. M. Matuszek, a nuclear chemist with considerable experience in all types of radiochemical and radiological health techniques. The principal contact for former Con-Rad customers at Isotopes, Inc., will be Mr. David Katzman.

Tracerlab Richmond Reports Half-Lives of ^{55}Fe and ^{196}Au

The Tracerlab Richmond Radiochemistry and Radiomeasurements Group has started to evaluate half-life data, some of which have been accumulating as much as 15 years. The first two values concern ^{55}Fe and ^{196}Au. A sample of ^{55}Fe has been counted 156 times since 1955 (approximately 4.5 half-lives) on an argon end-window detector. Through use of a linear regression analysis on an IBM-7094 computer, the data suggest a half-life of 2.79 ± 0.03 years. An isotopically purified source of ^{196}Au was counted on a NaI(Tl) detector for several half-lives. Linear regression analysis of the decay data gave a computed half-life value of 6.01 ± 0.06 days.

Chicago Studies Treatment of Sewage with Radiation

The Metropolitan Sanitary District of Greater Chicago is completing its plans to study the treatment of sewage by radiation, specifically to break down phenols and cyanides. Purdue Research Foundation and Battelle–Northwest Laboratories are under contract to perform the necessary research, and Dr. Albert Crewe has been retained as a consultant. Purdue will investigate the effect of radiation on solids under a 1-year contract, and Battelle will establish design criterions for treating liquid wastes under a 6-month contract. Federal grants totaling over $100 thousand have been requested to finance the project.

Officials of the Chicago Sanitary District said that a literature search failed to produce any significant references to treating industrial wastes with radiation.

World Food Panel Report Arouses Ire of JCAE, Army, and AEC

A special study of the world's food supply recently issued by a World Food Panel of the President's Science Advisory Committee contained the following paragraph:

> Radiation treatment to prolong storage life of perishable foods under refrigeration, to control insects in exported fruits and vegetables and stored grains, and to preserve perishables in hermetically sealed containers is technically possible, but not yet an efficient commercial reality. Recent studies suggest that harmful radiation effects may be transmitted by stable, toxic intermediate products in foods. The significance of these studies cannot be ascertained until extended feeding studies are completed. These and other problems indicate that radiation is not likely to have a significant application for food preservation in the foreseeable future.

The response was immediate. Letters were sent to the Office of Science and Technology, Office of the President, by John T. Conway, Executive Director of the staff of the Joint Committee on Atomic Energy of the Congress, and by A. A. Bertsch, Chairman, Interdepartmental Committee on Radiation Preservation of Food. Another letter came from the Department of the Army, and two letters came from the AEC—one from the General Manager's Office and one from Dr. Seaborg, Chairman of the AEC. All five letters took grave exception to the statement of the Food Panel and asked immediate clarification. Excerpts:

From Department of the Army, Col. R. T. Reid

The Army considers foods pasteurized or sterilized by irradiation under carefully controlled and specified conditions to be wholesome. ... Hence, the Army does not agree with the inference in the PSAC Panel's report ... suggesting that harmful effects may be transmitted by stable, toxic intermediate products in foods. ... In addition, the Army does not agree with the PSAC Panel statement in the same paragraph indicating that radiation is not likely to have a significant application for food preservation in the foreseeable future. ... The Army anticipates rapid progress within the United States in commercialization of irradiated foods, ...

From Dr. Seaborg's letter

AEC's view of potential applications is more optimistic. ... Extensive feeding studies ... have provided no evidence of harmful effects in animals fed large quantities of irradiated food for long periods of time and the FDA has cleared several irradiated foods for unlimited consumption. ... These considerations along with an ever increasing worldwide interest in developing the technological base required for effective application indicate that there is promise that the technique will find varied and significant applications in the foreseeable future and perhaps contribute to alleviation of world food shortages.

From John T. Conway's letter

The ... statement is contrary to the information on the use of radiation to preserve foods which has been supplied to the [JCAE]. Accordingly, please supply the Committee with an explanation of the basis of the statement in the panel report, covering in particular the details of the studies which are referred to. In addition, please inform the Committee of what reviews were made of the referenced paragraph by technical personnel responsible for food irradiation research under the Army's and Atomic Energy Commission's programs.

From A. A. Bertsch's letter

This evaluation appears to us to overlook many significant developments since the inception of the program and especially in recent years. Intensive wholesomeness studies on some 21 radiation sterilized foods conducted by the Surgeon General of the U. S. Army, and other studies on low dose radiation applications conducted by the Atomic Energy Commission, have shown irradiated foods to be wholesome. In fact, in a statement before the Joint Committee on Atomic Energy, June 9, 1965, the Surgeon General concluded that ... it can be stated that foods irradiated up to absorbed doses of 5.6 Megarads with a cobalt-60 source of gamma radiation or with electrons with energies up to 10 million electron volts have been found to be wholesome, i.e., safe and nutritionally adequate. ... The conclusion of your Subcommittee that radiation is not likely to have a significant application for food preservation in the foreseeable future overlooks such developments and may very well impede future progress here and abroad. ... It seems to us imperative that the implication in the report that this process is unsafe be clarified and that readers of the report have knowledge of the many favorable developments which suggest that the process may well have significant applications within the foreseeable future.

In response to these letters, Ivan L. Bennett, Deputy Director of the Office of Science and Technology, replied:

It is important to note that the paragraph is part of a section which addresses problems of marketing, processing, and distribution of food products in the *developing* countries where food shortages now exist and are worsening. ... In the same section of the Report, it is pointed out that procedures as well established as refrigeration and canning are inapplicable in all but very limited areas of the developing countries and, for example, that for about three-fourths of the population of India, there is no immediate prospect for the use of any processing method that will increase food costs. ... Food irradiation, a technology still under extensive research and development but not yet established on a commercial scale even in a developed country, is not believed by the Panel to be likely to contribute significantly to solution of the existing food problem in the developing countries. This by no means implies that the ongoing programs for industrial application in this country should be discontinued.

He went on to admit, in response to Conway's question, that:

... the paragraph was not reviewed by technical personnel responsible for food irradiation research under the Army or Atomic Energy programs.

Sales of Irradiation Plants May Depend on Food Irradiation

Rex Edwards, Managing Director of H. S. Marsh Ltd. in Britain, said at the dedication of Europe's first large food irradiator that the international market for large radiation process plants is dependent on the future of food irradiation. The Wageningen, Holland, plant is the sixth for Edwards; the other five are in Sweden, Italy, West Germany, and England. If only one developed country will approve for public sale one major irradiated food—such as fish in England, there will be a rapid market takeoff, according to Edwards. He thinks this is especially true of the underdeveloped countries, which have an urgent need for more and better food for their people.

Food-Irradiation Activities Abroad

At the 8th Japan Conference on Radioisotopes in Tokyo, Japanese officials revealed an 8-year $2-million food-irradiation program. The USSR is stepping up its sprout inhibition of potatoes, and Italy is nearing government approval of the process. The Israeli government has cleared irradiated potatoes. Canada is working on clearances for irradiation of strawberries and mushrooms; Canada is also pursuing further research on the elimination of *Salmonellae* in eggs and egg products. The Swedish and English

governments have begun work on obtaining clearances of radiation-pasteurized fish.

Douglas United Nuclear Drops Plutonium Fabrication Plant, Expands Other Facilities at Hanford

McDonald Douglas Corp. and United Nuclear Corp. have met with AEC officials and successfully argued that the plutonium fabrication plant, which was part of their 1965 bid to operate the Hanford facility as a joint company under the name Douglas United Nuclear, should be dropped. The three parties agreed that the plant—originally scheduled for 1967 operation and already postponed once—would not be needed before 1974. Douglas United Nuclear proposed definite alternatives that were realistic and helpful enough to dissuade AEC from canceling the contract entirely, which AEC did do to Isochem. McDonald Douglas will expand to $4 million its investment in the Donald W. Douglas Laboratories, which studies nuclear applications in space, and increase the staff of the laboratory to 170. United Nuclear will put further funds into the Zircaloy tube facility it shares with Sandvik Steel; this expansion will raise investment in the facility to about $4.4 million by 1970.

Neutron Radiography Newsletter Published Twice Yearly

The Society for Nondestructive Testing has been distributing an informal Neutron Radiography Newsletter twice a year, free to anyone who expresses an interest in receiving it. Each issue contains a bibliography, announcements, and a summary of radiography programs at various U. S. laboratories. It covers both Europe and the Far East. For copies, write Dr. Harold Berger, Building 212, Editor, Neutron Radiography Newsletter, Argonne National Laboratory, 9700 South Cass Avenue, Argonne, Ill. 60439.

Union Carbide Goes Ahead with Radiation-Treated Polyethylene Oxide

Union Carbide has named Parsons–Jurden to design and supervise construction of a plant to produce radiation-treated polyethylene oxide resin. The Charleston, W. Va., plant will treat the organic polymer to improve its viscosity by Union Carbide's patented ^{60}Co process. The facility will contain several kilocuries of ^{60}Co and is expected to be the largest commercial application of chemical process irradiation in the United States.

Smithsonian Institution Says Industrial Isotope Research Is $20 Million Per Year

Preliminary data received by the Smithsonian Institution's Science Information Exchange (SIE) indicate that industrial spending on isotopes and radiation research in the United States may be as high as $20 million per year. The SIE program is attempting to define the amount and type of nongovernment research; questionnaires were originally sent to about 500 companies, and a further questionnaire will be sent to 160 companies.

Australian Firm Acts for AECL

Consolidated Nucleonics Pty. Ltd. of Sydney, Australia, has become the exclusive agent for Atomic Energy of Canada Limited in that country. The new company was formed by Australian Consolidated Industries Ltd. and Swift and Company Ltd. The new company will provide competition for the government-produced ^{60}Co from the High Flux Australian Reactor.

NUMEC To Expand Radiation Processing

Nuclear Materials and Equipment Corp. (NUMEC) will soon acquire 100 thousand curies of ^{60}Co from Atomic Energy of Canada Limited, and an order for another 100 thousand curies will be up for bids by this summer. The two orders will be used with the present 40 thousand curies of ^{60}Co at the Quehanna, Pa., radiation processing plant of NUMEC.

The company is moving strongly into the radiation processing industry, mainly as a result of support from its parent company, Atlantic Richfield. Irradiated bacon will be test marketed soon, and NUMEC has already started test marketing of irradiated potatoes. Its main product will be PermaGrain* flooring, an irradiated wood–plastic material.

Recent Accessions of Isotopes Information Center†‡

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*Trademark of Nuclear Materials and Equipment Corp.

†International Atomic Energy Agency (IAEA) publications may be obtained from the National Agency for International Publications, Inc., 319 East 34th Street, New York, N. Y. 10016.

‡All CFSTI reports are available for $3.00 (full size) and $0.65 (microfiche) from Clearinghouse for Federal Scientific and Technical Information, National Bureau of Standards, U. S. Department of Commerce, Springfield, Va. 22151.

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